Doctoral Thesis

Evaluation Methodology on Heavy Metals Transport and Pollution for Environment Management in Urban River Basin in Developing Countries.

(開発途上国の都市河川流域における環境管理のための重金属輸送と汚染の評価方法論)

Saadia Bouragba

M. Eng.

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Kitami Institute of Technology, Japan

بسم الله الرحمن الرحيم

In the name of Allah, the all-merciful, the all-compassionate

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This thesis is the fulfillment of the requirement for the Ph.D. degree in Department of Cold Region, Environmental and Energy engineering of Kitami Institute of Technology, Japan. The thesis is deal with the modeling of heavy metals transport and pollution for environment management in urban river basin in developing countries.

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Abstract

In recent several decades, using of numerical computational models is getting more popular by planners, managers, engineers and scientists in different fields. Therefore, the assessment based on numerical computational models in water quality management seems promising.

The industrial activities have been caused severe damage to water ecosystems over time. The intensive industrial activities in urban area caused a significant increase in the quantities of hazardous materials, being released into rivers during wastewater disposal. The heavy metals are among the most prominent and danger pollutants resulting by the industrial activities. The monitoring of these pollutants in riverine systems is one of the most important water management challenges, because of their toxicity and their potential to accumulate and persist in the environment. Several studies related to the water quality modeling have been demonstrated the effectiveness of the hydrological models as powerful tools in prediction of heavy metal fate and transport in riverine systems.

The main objective of this thesis is evaluation methodology on heavy metals transport and pollution with the help of a distributed hydrological model (DHM), for environment management in urban river basins in developing countries.

The thesis addresses the evaluation of the effectiveness of Geophysical Flow Circulation (Geo-CIRC) model based on Object Oriented Design (OOD) in modeling of multiple heavy metals transport in Harrach River in Algeria which is severely polluted with various heavy metals originating from industrial activities, in order to use as a helpful monitoring tool in water quality management of rivers. As well, a simple approach for varied partition coefficient (K_{d}) modeling was proposed in order to enhance and increase the model accuracy for simulation of heavy metals concentrations in river sediments.

The results showed that the Geo-CIRC model was able to simulate simultaneously

the transport of multiple heavy metals in a river and present a comprehensive description of river contamination with only a minimal amount of observational data, where the application of OOD increased the model's effectiveness, by improving the model's flexibility even many unknown point sources exist, and supported the inclusion of multiple heavy metals in the simulation with reasonable accuracy. Likewise, the proposed empirical multivariate regression model was useful in estimation of the potential variables of the partition coefficient with physicochemical properties changes, as well introducing these changes in simulation increased the result accuracy of the model.

Including of Geo-CIRC model in monitoring strategy can provide comprehensive assessment of the environmental state in the river systems with less cost in short time by using less effort because of the OOD's advantage. Likewise, the model feature allows us to be able to simultaneously utilize much information which could be taken from limited monitoring data. As well, the proposed partition coefficient model can enforce the availability of sediment data.

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Abbreviations

u_i^n	velocity vector at grid *i* and time *n*,
i	*Grid*
n	*Time*
g	gravitational acceleration
h_i	depth at grid *i*
A_i	cross-sectional area at grid *i*,
Δt	time step
Z_b	distance from datum level to bottom boundary,
n_m	Manning's roughness coefficient
q_L	Lateral inflow.
C	concentration of heavy metals dissolved in water column,
Q	Discharge
A	wetted cross-sectional area
A_a	source or sink of dissolved heavy metal,
B_a	transformation flux from, or to, adsorbed particulate phase onto the sediment,
D_{tx}	diffusion coefficient
Q	lateral inflow or outflow discharge
C_a	lateral inflow or outflow dissolved heavy metal concentration,
Δx	Distance between two consecutive cross-section which can be either constant or variable.
SP	concentration of heavy metals in sediment,
A_{SPa}	source or sink of absorbed particulate heavy metal

B_{SPa}	Transformation flux from, or to, adsorbed particulate phase onto the sediment.
C_T	total concentration
K_d	partition coefficient
Km^2	Kilometer squared
%	Percent
mg/L	Milligram per liter
mg/kg	Milligram per kilogram
m	Meter
s	Second
m^3	Meter cubic
mm	Millimeter
Pb	Lead
Hg	Mercury
Cr	Chromium
Zn	Zinc
Cd	Cadmium
SS	Suspended solid concentration
pH	
BOD	Biological oxygen demand
COD	Chemical oxygen demand
EC	Electric conductivity
1D	One dimension
ADE	Advection dispersion equation
CBR	Case-based reasoning
DHM	Distributed hydrological model
DEM	Digital elevation model

EFDC	Environmental Fluid Dynamics Code
Geo-CIRC	Geophysical flow Circulation
HEC-HMS	Hydrologic Engineering Center Hydrologic Modeling System
OOD	Object-Oriented Design
RBF	Radial Basis Function
TREX	Two-dimensional, Runoff, Erosion, and Export
WASP	Water Quality Analysis Simulation Program
JICA	Japanese international cooperation agency
ONEDD	Is the national observatory of the environment and sustainable development in Algeria
ONM	Is the national meteorological office in Algeria
RGHP	Is the general census of population and housing in Algeria

General introduction

1. Background

The contamination of aquatic system with heavy metals is posing a major risk to the public health because of the toxicity of these pollutants, as well their environmental persistence and the ability to incorporate into food chains. The pollutants can enter aquatic systems through different ways, whereas the industrial activities effluents are the most prominent source [1], [2].

Many rivers in worldwide are suffering the problem of heavy metals pollution due to the wastewater discharged from industrial activities processes over several decades. Harrach River in Algeria represents a striking example of heavy metal pollution caused by the effluents of industrial activities. The inadequate sewage systems and the discharge of untreated industrial wastewater into the river are the principal contributed to the exposure of this river to pollution problem. Arguably, the bad water quality management planning and the weak in the enforcement of environmental laws are also significantly contributed to increase the pollution level in this river [3].

In recent several decades, the numerical models have proven their effectiveness and good performance in predicting the transport of different pollutants in environmental systems. Therefore, the assessment based on numerical computational models is getting more and more popular by the water resource planners, water quality managers, engineers and scientists [4], because of operational performance and low cost of numerical as monitoring tools [5]. Also, through the numerical modeling, we can visualize the fate of pollutants entering the riverine system by measuring their interactions and transport. Furthermore, the modeling process can give an estimation of required values of pollutant levels at the locations where the observation data are not available.

Many studies in the field of water quality modeling were carried out in order to understand the dynamics of the transport of pollutants such as heavy metals in rivers. Where, the hydrological models have been used in the simulation of the behavior and transport of heavy metals in riverine system. For example, Falconer *et al.* (2005) [6] and Kashefipour & Roshanfekr (2012) [7] studied the different processes involved in the transport and distribution of pollutants in rivers and estuaries using hydrological models, and they proposed a conceptual framework which would help and support the application of these models in water quality management.

It should be noted that in the studies related on heavy metal transport modeling, the simulation of metals transport processes in aquatic systems is usually considered separately for each element as well reactions. Also, the calibration of models requires a large numbers of observations, which are very limited especially in developing countries. In study carried out by Bouragba *et al.* (2017) [8], Geo-CIRC model based on OOD originally developed by Nakayama *et al*. (2015) [9], could be applied successfully to estimate lead (Pb) and mercury (Hg) concentrations in stream water and sediment of Harrach River in Algeria based on only a few observations data. The GeoCIRC model used in this research, can analyze the interaction among essential hydrological processes, such as surface water flow, river flow, infiltration layer flow and groundwater flow.

The partitioning of heavy metals between particulate and water phases in polluted aquatic system is intricacy phenomenon that is strongly related to the environmental conditions. The partition coefficient (K_d) is an empirical parameter which depends on various factors, and it is commonly used for describing solid-solution interaction [10]. Therefore, the incorporating of the metal K_d by considering the environmental conditions changes in heavy metals simulation, seems promising for the improvement and enhance of the model accuracy.

2. Objectives and scope

The monitoring of heavy metal transport originating from wastewater is important as one of the mains depart point of water quality management of river basins in urban areas, because the urban river is the principal receiver of the different wastewater (urban, industrial, etc.) which are a source of various pollutants. The main objective of this thesis is assessing and improving the performance of a distributed hydrological model in order to evaluate a methodology on heavy metal transport and pollution for environmental management in the urban rivers in developing countries (Case study: Harrach River in Algeria).

- At the first stage, simulation results in Harrach River in four elements of heavy metals (namely: Pb, Hg, Cr and Zn) were validated by using historical data. Also, assessment on the model accuracy and efficient water monitoring plan was discussed based on different simulation cases controlling pollutant sources.
- In the second stage, an empirical multivariate regression model of K_d considering physicochemical properties (pH, suspended solid concentration (SS), and organic content (OM)) in riverine water is proposed, then, the concentrations of lead (Pb) in sediment were simulated by using the numerical model incorporating with the K_d model in order to improve the accuracy of simulations results.
- In the final stage, we will try to present the benefices of using Geo-CIRC model in monitoring system, in order to highlight the practical outcomes of this study.

3. Thesis outline

The thesis is consisting of five chapters:

- **Chapter 1** summarizes a literature review. This chapter consist of three subtitles:
    - *Heavy metals*; where we addressed the following points: Definition and properties, Origin, Toxicity, Impact of heavy metal contamination in aquatic ecosystems;
    - *Harrach River*: in this part we addressed the following points: Location and description, Problem of pollution, Water quality management in Algeria
    - *Water quality modelling:* we have addressed the following points: Description, Studies and application, Heavy metal transport in river system
- **Chapter 2** provides the detail of evaluation of the performance of the Geo-CIRC model based on OOD for the simulation of contamination from various heavy metals (i.e. Pb, Hg, Cr, and Zn) in Harrach River in Algeria. A general overview and methods are described. Then the results are extensively discussed. Finally, finding conclusion is presented.

- **Chapter 3** presents the detail of a simple approach modeling of heavy metals concentrations in sediment, undertaken the using of K_d model considering various physicochemical properties (pH, SS, biological oxygen demand (BOD) and chemical oxygen demand (COD)).

    - First, the generation of regression models of K_d values based on pH, SS, BOD and COD is discussed.
    - Then the simulation of Pb concentrations in sediment undertaken varying K_d is also described. Finally, the model performance was evaluated.

- **Chapter 4** presents the benefices of using Geo-CIRC model in monitoring system, in order to highlight the practical outcomes of this study.

- Finally, **Chapter 5** summarizes the findings and concludes the thesis.

Chapter 1

Literature review

1.1 Heavy metals

1.1.1 Definition and properties

"Heavy metal" is common term refers to any dense metallic elements those are toxic or poisonous at low concentrations [1]. They are stable elements of high specific gravity and atomic weight. As well, they characterized by luster, ductility, malleability, and high electric and thermal conductivity.

1.1.2 Origin

Heavy metals in nature are existed from weathering of minerals, erosion and volcanic activity, and it can also originate from the anthropogenic activities such as industrial and urban activities (**Fig.1.1**).

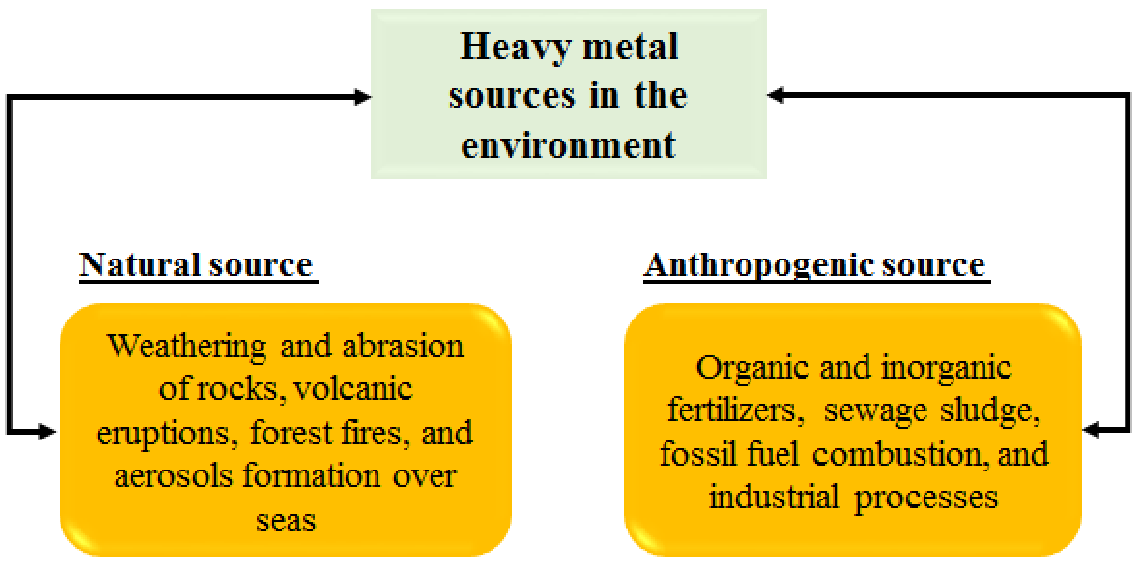

Fig.1.1 Heavy metal sources in environment.

The physicochemical forms of heavy metals are significantly influencing on their bioavailability, toxicity, transport, and fate in environment (water, soil, and sediment) [2], [11].

1.1.3 Toxicity

The heavy metals are generally non-conservative substances in nature and they cannot be degraded or destroyed. They enter our bodies via food, drinking water and air, and they are essential in many human needs. Some heavy metals (e.g. copper, selenium, or zinc) are indispensable essential elements to maintain human metabolism [12], [13]. Moreover, they are of outstanding technological significance, e.g., iron, zinc, tin, lead, copper, tungsten, etc. Not to mention among them the noble elements such as gold, silver, iridium, rhodium, or platinum. However, many of these element, e.g., mercury, cadmium, arsenic, chromium, thallium, lead, and others, represent the "dark side of chemistry" because they have toxic effects even at low concentrations [13].

The heavy metals pose a major threat as several health risks are associated with their toxicity. Whereas, their toxicity causes malfunctioning of the body system. These elements also sometimes act as pseudo elements of the body, interfering with the metabolic processes that occur in the body resulting into chronic diseases [14].

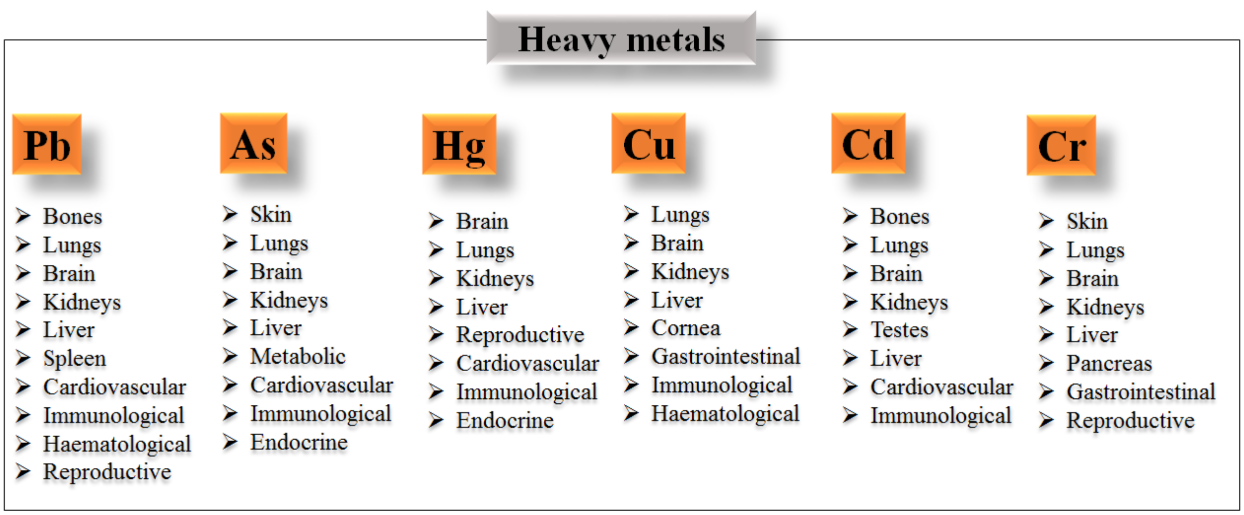

Fig.1.2 Impact of some heavy metals.

1.1.4 Heavy metal contamination in aquatic ecosystems

The heavy metals are considered as important contaminants of aquatic environments worldwide [15]. Usually, the heavy metal in aquatic systems exist either dissolved in water or suspended in particles to finally settle down at the bottom or taken up by the organisms. Heavy metal in contaminated aquatic system have the ability to accumulate in aquatic flora and fauna, which are in turn enter into human food chain (**Fig.1.3**), thus representing a threat to the human health [16].

The progressive and irreversible accumulation of heavy metals in various organs of aquatic organisms leads to different diseases in the long run because of their toxicity, therefore exposed the aquatic biota and other organisms to a serious risk [17]. The toxicity of metals in the aquatic environment is influencing by various factors such as speciation, solubility and complexation of metals. In addition, heavy metal interaction can change their toxicity effects on organisms living in aquatic system even positively or negatively [15].

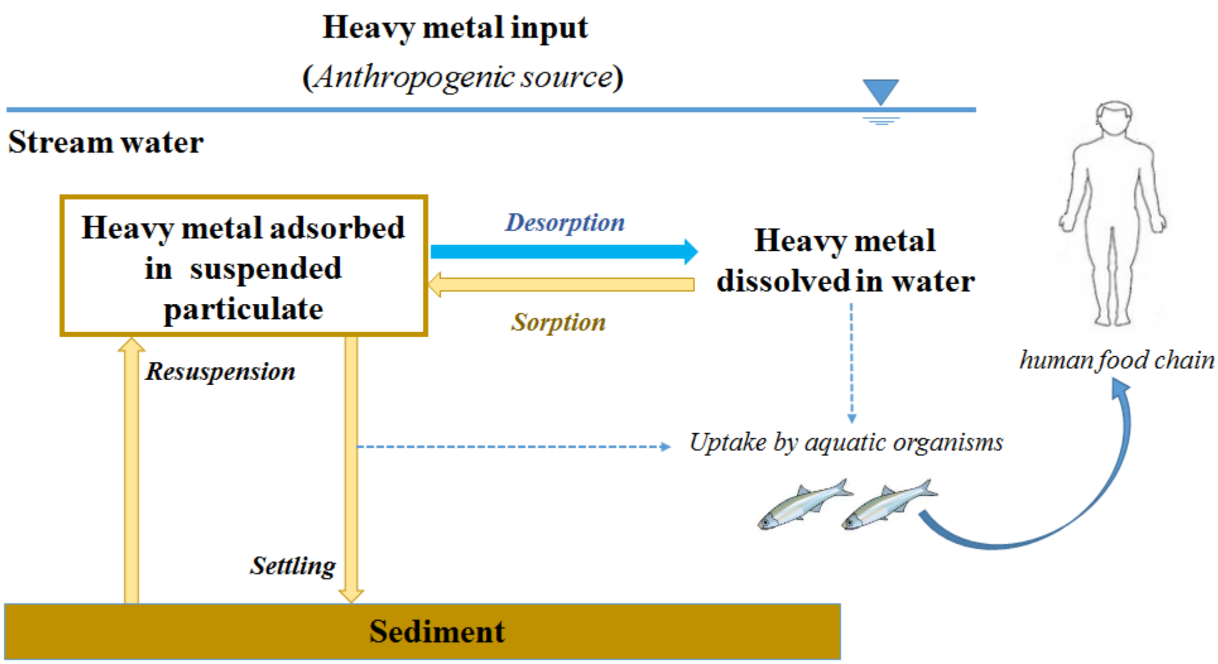

Fig.1.3 Heavy metal in aquatic environment.

1.2 Harrach River basin

1.2.1 Location and description

Harrach River Basin is located in north of Algeria (**Fig.1.4**). This river is considering as one of the large rivers, extends overs an area of 1270 km^2 [3]. Harrach River has provided important water resources in Algiers, the capital city of Algeria. The water resource in this river is feed by rainwater, river water from its tributaries, surface runoff, urban wastewater and industrial wastewater. The average discharge of Harrach River is ranged from 4 to 5 m^3/s but it may change to zero during the dried period and to 3000 m^3/s during the period of inundation [18].

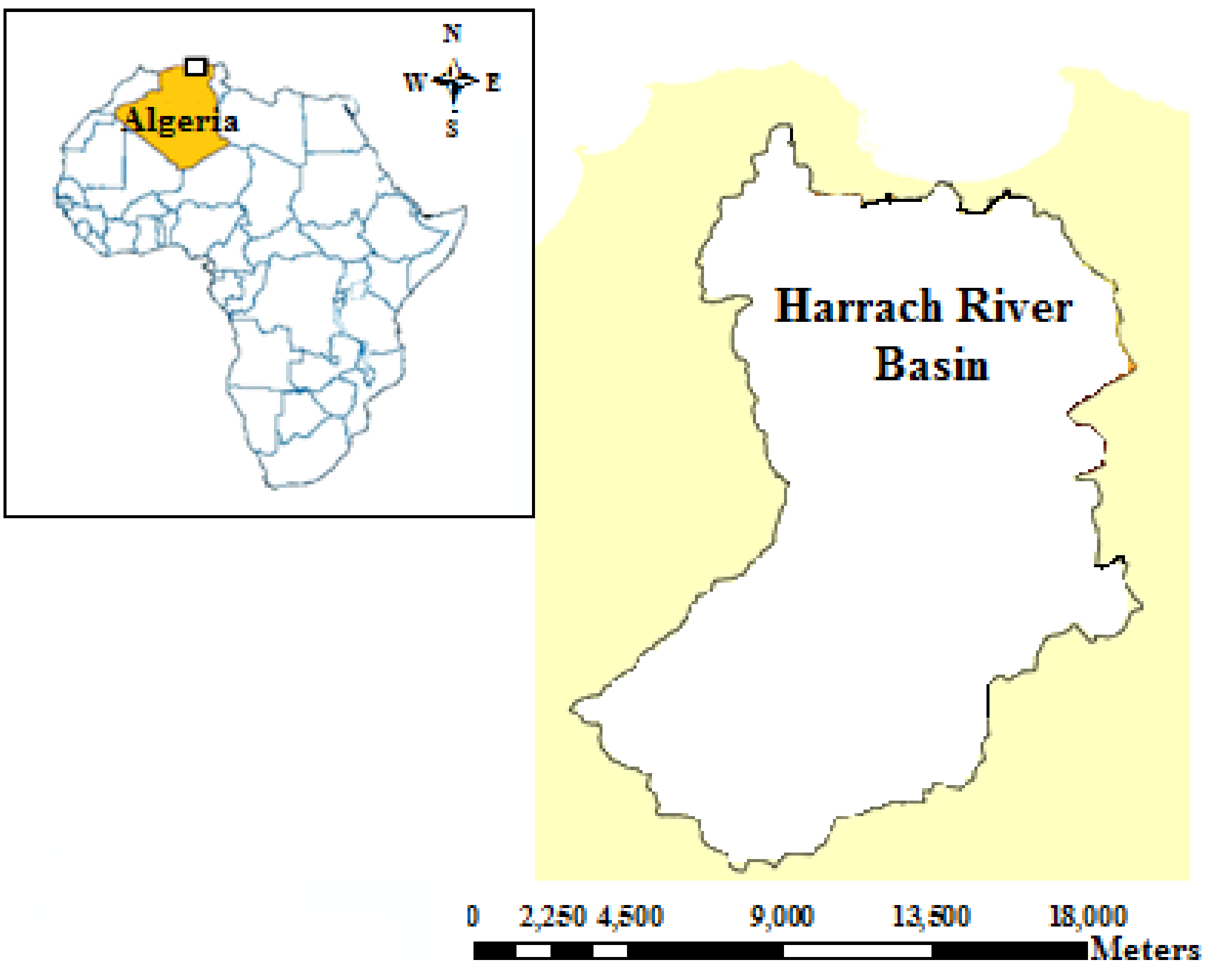

Fig.1.4 Harrach River basin

1.2.2 Climate

All of the North of Algeria is almost Mediterranean climate with a mild and wet climate in winter, hot and dry in summer. Altitude, position and exposition lead to tremendous differences between regions, continental characteristics are combined quickly when we advance towards the interior with Mediterranean traits [3].

Generally, the rains are irregular and sometimes unequally distributed. In summer precipitations are very scanty, they reach to the maximum (abundant) in the Tell Atlas in winter, and in the high plains in spring [3]. The study area in this thesis, is governed by a moderate Mediterranean climate characterized by the alternation of the hot dry season and a wet season, rainy and relatively cold. The temperature rarely exceeds 40 °C and almost never drops below 0 °C [18]. The annual rainfall average is around 805 mm (year- 2010) according to the national office meteorology in Algeria (ONM: Office National Météorologique).

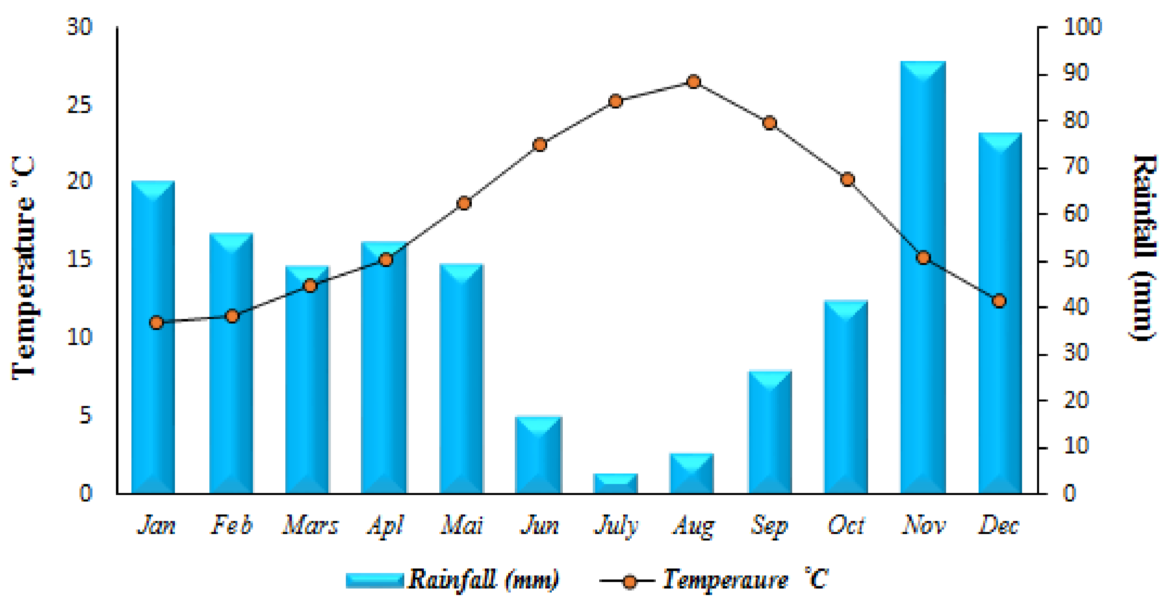

Fig.1.5 Variation of the temperature and rainfall (ONM.2010).

1.2.3 Urbanization and population

In 2014, the population in Algeria has reached 38.7 million inhabitants according to RGHP (Recensement Génaral de la Population et de l'Habitat), with 2.16% of population growth rate. The distribution of population in Algeria territory is unbalanced, where, 63% of population is grouped in north (4% of territory), following by 28% in the highlands (9% of territory), while the south covers only 9% of the population (87% of territory). This unbalanced distribution is due to the development and economical changes in the country that resulting overpopulation in big cities located in the north [19].

1.2.4 Problem of pollution in Harrach River

After the political independence in 1962, Algeria witnessed a significant economic development and benefitted from a large local and foreign investment. In the 1980s, Algeria was considered as the largest emerging industrialized country in Africa. At that time, a significant growth of industrial activities operation in various sectors (about 240 mega-scale industrial units). The most of these activities is potential sources of various pollutants such as organic compounds, heavy metals, petrochemicals, and other toxic substances that require appropriate pollution control or treatment facilities [20].

A large industrial zones are spread around Harrach River basin (**Fig.1.6**). Unfortunately, the most of the activities in these zones are operating without any environmental controls, causing pollution problems of the river with various pollutants [3].

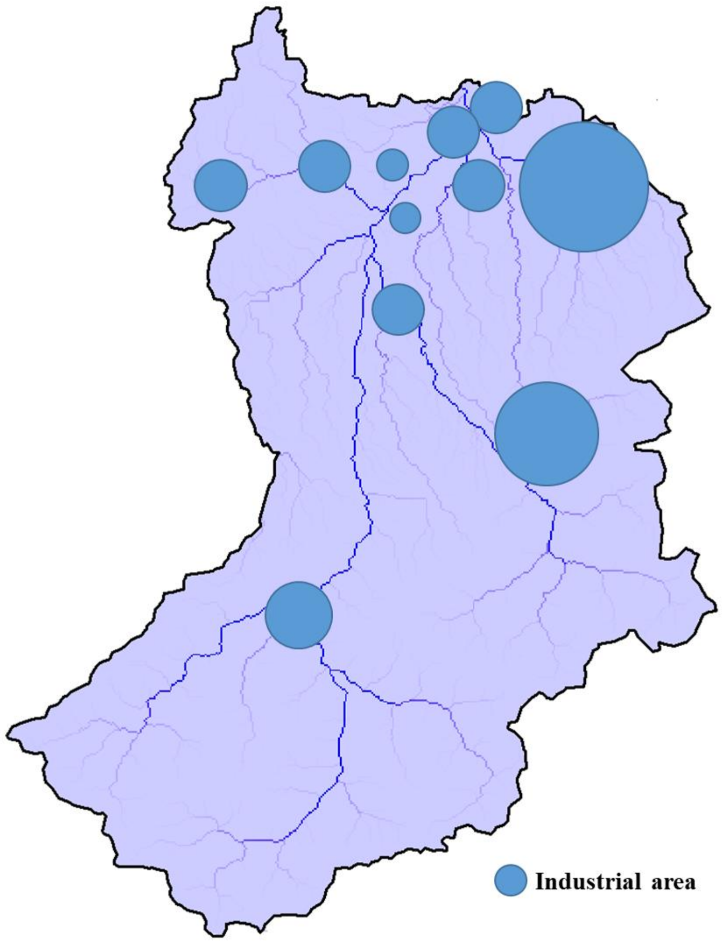

Fig.1.6 Industrial zones location around Harrach River basin.

1.2.5 Problem of heavy metal pollution Harrach River

The assessment study carried out by the cooperation team ONEDD/JICA (Observatoire National de l'Environnement et de Developpement Durable (ONEDD) and the International Cooperation Agency of Japan (JICA)) in order to determine the levels of different heavy metals concentrations in Harrach River and its tributaries, from 2004 to 2011. The study found a high levels of various heavy metals concentrations in most of the sampling locations along the Harrach River Basin. These high levels are caused by the wastewater resulting from the major industrial activity and discharging directly into the river without any treatment [21].

1.3 Modeling

1.3.1 Description

According to Anu M. (1997) [22], modeling is the process of making a model, in order to predict the impact of changes on a system. The model represents the construction and working of a system, where the model is similar to the system but simpler.

In modeling process, it should first collect and prepare of input data, as well evaluate the necessary parameters for the model setup, and prepare for the model implementation. In the second step, the model is evaluated if it has achieved the required purpose, and that is through calibration and validation, as well, post-audit. Finally, the final use of the model.

1.3.2 Model Selection

The selection of model can be stated based on following simple guidelines [23]:

- Define the problem and determine what information is needed and what questions need to be answered.
- Use the simplest method that will yield adequate accuracy and provide the answer to your questions.
- Select a model that fits the problem rather than trying to fit the problem to a model.
- Do not forget the assumptions underlying the model used and do not read more significance into the simulation results than is actually there.
- The cost of maintaining and updating the model over time must be acceptable.

1.3.3 Water quality modeling

Water quality predictive models include both mathematical expressions and expert scientific judgement. Besides that, this models are based on process-based (mechanism) models and data-based (statistical data) models [23]:

- The models should link management options to significant response variables such as pollutant sources and water quality standard parameters (pH, EC., salinity ...etc.).
- Process-based models should be consistent with scientific theory.

- It should be reported the model prediction which requires the prediction error estimates to provide decision makers with estimates of the risks of options.
- The models selected should be appropriate with complexity of the situation and to the available data.
- It should be not use the models requiring large amounts of monitoring data when this data is unavailable. Beside the possibility of updating and improving the model as appropriate [24].

1.3.3.1 Modeling Approaches

According to Nirmala Kh. N., (2002) [25], the most common modeling approaches in the environmental area can be classified into three basic types:

- Physical modeling or “experimental modeling”, includes the representation of the real system by a geometrically and dynamically similar, scaled model and making observations and measurements by conducting experiments on it. These observations and measurements from experiments are extrapolated to the real systems.
- Empirical modeling or “black box modeling”, is based on empirical observations rather than on mathematically describable relationships of the system modelled. In this model the results are considered a "black box" reflecting only what changes could be expected in the system performance due to changes in inputs.
- Mathematical modeling or “Mechanistic modeling”, is based on theoretical approach where the mathematical relations are derived between the variables known to be significant by using the principles and fundamental theories which govern the system with simplifying assumptions. The historical data from the real system is used to calibrate the model results and using additional data to validate it, then can be made predictions with predefined confidence. The mathematical model is reflecting how changes in input are affected the changes in system performance.

1.3.3.2 Water quality modeling application

The use of mathematical models has become widespread in many fields, particularly in environmental fields. Several studies related to the water quality modeling have been reviewed the processes involved in the transport and behavior of contaminants in aquatic systems in several aspects, i.e., distribution, reactions, and monitoring methods, and provided results which are helpful in illustrating various aspects in water quality modeling field, for example we mention:

Pak *et al.* (2015) [26] used EFDC model (Environmental Fluid Dynamics Code) to simulate of discharge and TSS (total suspended solid concentrations) in upstream and downstream of the Baekje Weir installed in Geum River, Korea, in order to consider the characteristics of changes in sediment transport being result of weir installation

Zhang *et al.* (2017) [27] have effectively predicted the biochemical oxygen demand (BOD) in the sewage treatment, by using integrated model combining of the Radial Basis Function (RBF) neural network and improved case-based reasoning (CBR). A high measuring accuracy is showed in the experimental results.

Gao *et al.* (2017) [28] have applied HEC-HMS (Hydrologic Engineering Center Hydrologic Modeling System) for the simulation of basin runoff and for the examination the urban agglomeration polders impact on the flood events in Qinhuai River, China.

Kachiashvilia *et al.* (2006) [29] came up with a mathematical model for simulating diffusion and transport of chemicals in rivers. Finite difference approximation and appropriate numerical algorithms were used for these models. The predicted results of the concentrations of polluting substances in the rivers matched well with the real data.

Md.Jahangir *et al*. (2008) [30], evaluated the performance of DHM in predicting nutrient concentration in river basin. The model has produced a good accuracy in simulation of nutrient concentration transport in Saru River basin in Japan.

1.3.3.3 Heavy metal transport in river system

In the field of heavy metals transport modeling, many studies have been conducted on the behavior and transport of these pollutants in riverine systems.

Florimond *et al.*, (1998) [31] applied WASP (Water Quality Analysis Simulation Program) to simulate the distribution of various heavy metals in Scheldt estuary, Belgium, under average hydrodynamic and SS sediment transport regimes. The result simulation of the distribution of the sorbed heavy metals, suspended sediment and salinity were agreed well with observation results.

Mark L.*et al.* (2008) [32] developed (Two-dimensional, Runoff, Erosion, and Export) TREX model, to simulate chemical substance including heavy metals transport and fate in the watershed scale, this model is an appropriate tool for investigating multimedia environmental problems that involves water, soils and chemical interactions in a spatially distributed manner within a watershed.

Roshanfekr *et al.* (2008) [33] were used a hydro-environmental model to predict the dissolved Pb and Cd concentrations along rivers introducing a new approach of the varied reaction coefficients used in the advection–dispersion equation (ADE)). The simulation results were successfully compared with the corresponding observed values.

1.3.3.3.1 Heavy metal transport modeling

1. The transport of heavy metals in the water can be described by 1 D advection – dispersion equation (ADE) [7]:

$$\frac{\partial CA}{\partial t} + \frac{\partial CQ}{\partial x} - \frac{\partial}{\partial x}\left[AD_{tx}\frac{\partial C}{\partial x}\right] = A(A_a + B_a) \quad (1.1)$$

Where C = concentration of heavy metals dissolved in water column, Q = discharge, A = wetted cross-sectional area, A_a = source or sink of dissolved heavy metal, B_a = transformation flux from, or to, adsorbed particulate phase onto the sediment, D_{tx} = diffusion coefficient.

Sources or sinks of dissolved heavy metals can be defined as [7]:

$$A_a = \frac{QC_a}{\Delta x} \tag{1.2}$$

Where Q = lateral inflow or outflow discharge, C_a = lateral inflow or outflow dissolved heavy metal concentration, Δx= distance between two consecutive cross-sections which can be either constant or variable.

The transport of adsorbed particulate heavy metal can be described by the 1D advective-dispersion equation [34]:

$$\frac{\partial SPA}{\partial t} + \frac{\partial SPQ}{\partial x}\frac{\partial}{\partial x}\left[AD_{tx}\frac{\partial SP}{\partial x}\right] = A(A_{SPa} + B_{SPa}) \tag{1.3}$$

Where SP = concentration of heavy metals in sediment, Q = discharge, A = wetted cross-sectional area, A_{SPa} = source or sink of absorbed particulate heavy metal, B_{SPa} = transformation flux from, or to, adsorbed particulate phase onto the sediment.

Heavy metal the distribution between water and particulate phases can be described by partition coefficient (K_d) [34].

The partition coefficient K_d is an empirical parameter which depends on various factors, and it is commonly used for describing solid-solution interaction [35]. The partition coefficient of heavy metal is simplified by assuming that the concentration of the metal sorbed to the solid particle is proportional to the concentration of the metal in solution [10] is expressed as

$$K_d = \frac{C_{sed}}{C} \tag{1.4}$$

Where C = concentration of heavy metals dissolved in water column, C_{sed} = concentration of heavy metals in sediment.

The concentration of heavy metal in sediment can be derived from equation (1.4) as following:

$$C_{sed} = K_d \times C \tag{1.5}$$

Chapter 2

Assessment of distributed hydrological model performance for simulation of multi-heavy metal transport in Harrach River, Algeria

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2.1 Introduction

The rivers in urban area are severely suffer with severe damage due to the intensive industrial activities. That's because the industrial activities in these areas causes an increase in the quantities of various pollutant such as heavy metals in aquatic system through their wastewater, in absence of adequate treatment facilities, or not properly operating.

In Harrach River in Algeria, the industrial activities are considering as the major source of pollution of this river with different pollutant. According to Yoshida *et al*, (2005) [21], the concentration of various heavy metals at the downstream of Harrach River is significantly high causing by the untreated wastewater that the industrial factories discharged directly into the river.

The monitoring of heavy metals in rivers based on simple water quality measurement methods needs time and labor-intensive besides the high costs. Whereas, the attractiveness of numerical models as powerful monitoring tools highlights by the ease of use, accessibility and low cost of computer systems and software. Therefore, using of numerical models can give an opportunity to improve the performance of the monitoring methods with less cost [5], [6].

In the context of water quality modeling, the hydrological models have been widely used by many researchers to predict the behavior and the transport of heavy metals in aquatic systems.

For example, the methodology provided by Kashefipour & Roshanfekr (2012) [7] predicted dissolved concentrations of Pb and Zn using a varied reaction coefficient approach to the source term of the Advection-Dispersion Equation. Likewise, they assessed the impact of pH and EC on the reaction coefficient in order to introduce the best relationships for reaction coefficients in term of pH and EC to improve the model accuracy.

Falconer & Lin (2003) [27] used hydro-environmental models to simulate the distribution of Cd and Zn concentrations along the Mersey basin, UK, with 3-D advective–diffusion equation and dynamic partitioning coefficients being used to link the metal concentrations and sediments. The simulation results were in good agreement with corresponding measured data.

In most of previous studies related to the modeling of heavy metals transport in rivers, the researchers are considering the transport process of each element in each advection-dispersion equation, separately. As well, a large number of observations are required the model's calibration.

Bouragba *et al*, (2017) [8], has been use the Geo-CIRC model based on OOD to estimate Pb Hg concentrations in Harrach River in Algeria. This model is originally developed by Nakayama *et al.*, (2015) [9], and it was successfully applied to estimate the metal concentrations in water and sediment based on only a few observations [8]. The characteristic of inherit in object-oriented approach allows the extension to include a new scheme in the model, which already exist in the module, and increase the model flexibility without changing the fundamental structure [37]. As well, the OOD allows the model to handle multiple transportable objects. Thus, the incorporation of multiple heavy metals in the simulation would contribute to the assessment of the point source impact in different elements simultaneously even with a few numbers of observations.

In this chapter, four heavy metals (namely: Pb, Hg, Cr and Zn) transport in Harrach River were simulated by using the Geo-CIRC model based on OOD. The model performance was assessed in order to investigate new strategies for monitoring of heavy metals contamination in rivers in developing countries, where the observational data limited.

2.2 Methods

2.2.1 Case study area

The study area is located in Harrach River Basin (**Fig.2.1**). This area has been chosen based on the outcomes of evaluation study carried out by the cooperation team ONEDD/JICA on Harrach river pollution with various heavy metals caused by the industrial activities. Which revealed a high level of various heavy metals such as Pb, Hg, Cr and Zn in sediment and water at different points along the river network [21].

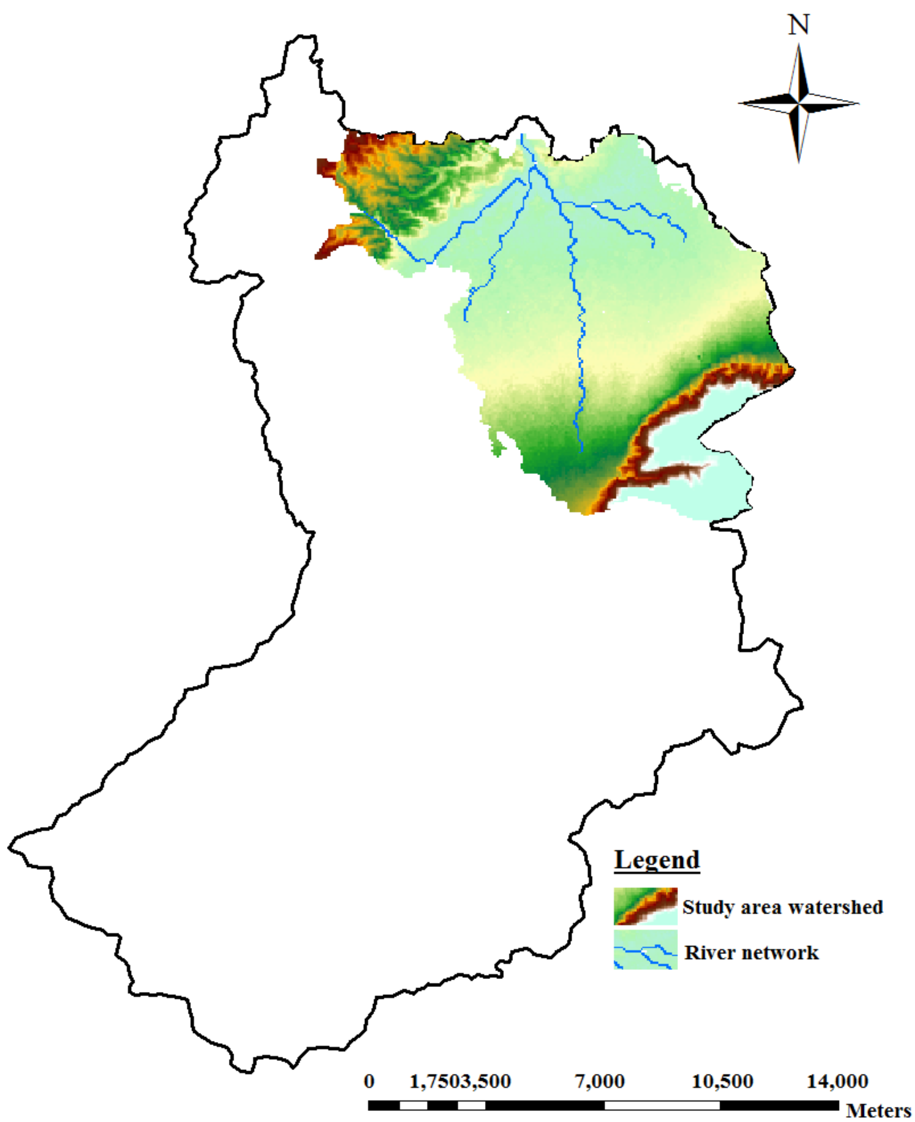


Fig.2.1 Study area location.

2.2.2 Model formulation

In this study, Geo-CIRC model has been applied for the simulation of Pb, Hg, Cr, and Zn concentrations. Based on physical processes this numerical model can simulate all the flow regimes such as water flow, river flow, infiltration flow and groundwater flow, and analyze the interaction between essential hydrological processes [8].

2.2.2.1 River flow model

The longwave equations are applied to estimate water level and discharge. The predictor-corrector method proposed by Nakayama *et al*. (2000) [38] was used, as in equations (2.1a) to (2.1e).

The validity and the applicability of the numerical scheme was verified from comparisons with the fully-nonlinear and strongly-dispersive internal wave equations [39], [40], [41]. One of the most significant characteristics of the Geo-CIRC model is that all branch rivers are connected to the main river as lateral inflows, which enables the reproduction of progressive longwaves up-stream without any discontinuity in the momentum (equation (2c)).

$$\frac{\tilde{u}_i - \hat{u}_i^n}{\Delta t} + \left[u\frac{\partial u}{\partial x}\right]^n = 0 \tag{2.1a}$$

$$\frac{\tilde{u}_i - \hat{u}_i^n}{\Delta t} = -g\nabla\left(h^n + Z_b\right) - \frac{g n_m^2 \hat{u}_i \left|\hat{u}_i\right|}{(h)^{4/3}} \tag{2.1b}$$

$$\frac{u_i^{n+1} - \tilde{u}_i}{\Delta t} + g\nabla\phi_h = q_L \tag{2.1c}$$

$$\phi_h = h_i^{n+1} - h_i^n$$

$$\frac{A_i^{n+1} - A_i^n}{\Delta t} + \frac{\partial}{\partial x}\left(A^n + \phi_A\right)\left(\tilde{u}_i + \Delta t g \frac{\partial}{\partial x}\phi_h\right) = 0 \tag{2.1d}$$

$$\phi_A = A_i^{n+1} - A_i^n \tag{2.1e}$$

where u_i^n = velocity vector at grid *i* and time *n*, *g* = gravitational acceleration h_i = depth at grid *i*, A_i = cross-sectional area at grid *i*, Δt = time step, Z_b = distance from datum level to bottom boundary, n_m = Manning's roughness coefficient which is estimated as 0.04 in the present simulation, and q_L = lateral inflow.

2.2.2.2 Heavy metal model

The distribution of heavy metals concentrations dissolved in water and adsorbed in particulate can be modeled by the advection equation [7].

The 1D advection equation for simulation of dissolved heavy metal concentration is described as following:

$$\frac{\partial CA}{\partial t} + \frac{\partial CQ}{\partial x} = A(A_a + B_a) \quad (2.2)$$

where C = dissolved heavy metal concentration, Q = discharge, A = wetted cross-sectional area, A_a = source/sink of dissolved heavy metal, and B_a = transformation flux from, or to, adsorbed particulate phase onto the sediment.

Sources/sinks of dissolved heavy metals can be estimated using the following equation [7]:

$$A_a = \frac{q_L C_a}{\Delta x} \quad (2.3)$$

where C_a = lateral inflow of heavy metal concentration, Δx = distance between two consecutive cross-sections which can be either constant or variable.

The transport of adsorbed particulate heavy metal is described by the following equation:

$$\frac{\partial SPA}{\partial t} + \frac{\partial SPQ}{\partial x} = A(A_{SPa} + B_{SPa}) \quad (2.4)$$

where SP = heavy metals concentration adsorbed in particulate, A = wetted cross-sectional area, A_{SPa} = source or sink of adsorbed particulate heavy metal, and B_{SPa} = transformation flux from, or to, the adsorbed particulate phase onto the sediment.

The total concentration of heavy metal in stream water can be estimated by summing the dissolved heavy metal and the adsorbed particulate heavy metal.

$$C_T = C + SP \quad (2.5)$$

where C_T = total concentration, SP = concentration of heavy metals adsorbed in particulate, and C = concentration of heavy metals dissolved in the water column.

Transport of total heavy in water can be described as following:

$$\frac{\partial C_T A}{\partial t} + \frac{\partial C_T Q}{\partial t} = A\left[\left(A_a + A_{SP}\right) + \left(B_a + B_{SP}\right)\right] \quad (2.6)$$

Noting that: $B_a = - B_{Spa}$

Equation (2.6) becomes:

$$\frac{\partial C_T A}{\partial t} + \frac{\partial C_T Q}{\partial t} = A\left(A_a + A_{SP}\right) \tag{2.6.a}$$

The distribution of heavy metals between the dissolved and adsorbed particulate phases can be described by the partition coefficient [35].

$$K_d = \frac{SP}{C} \tag{2.7}$$

From equations (2.5) and (2.7), the dissolved heavy metal can be expressed as follows:

$$C = \frac{C_T}{1 + K_d} \tag{2.7a}$$

In present study, the interaction of the heavy metal concentration between water and particulate phases was assumed to be in an equilibrium state which is expressed in the equation (2.7a).

2.2.3 Data preparation

2.2.3.1 Watershed and river network generation

The watershed and river network in study area were generated by using ArcGIS, in order resolve surface topography and the spatial distribution of point sources pollution. Digital elevation model (DEM) data obtained from the Consortium for special information (CGIAR-CSI) [42] were used to calculate the river network. The resultant rectangular raster grid has 246 columns and 201 rows. The delineated river network comprises 5 branches numbered from 0 to 4 (**Fig.2.2**). The watershed and the river network were simulated with a surface grid of 100 m cells.

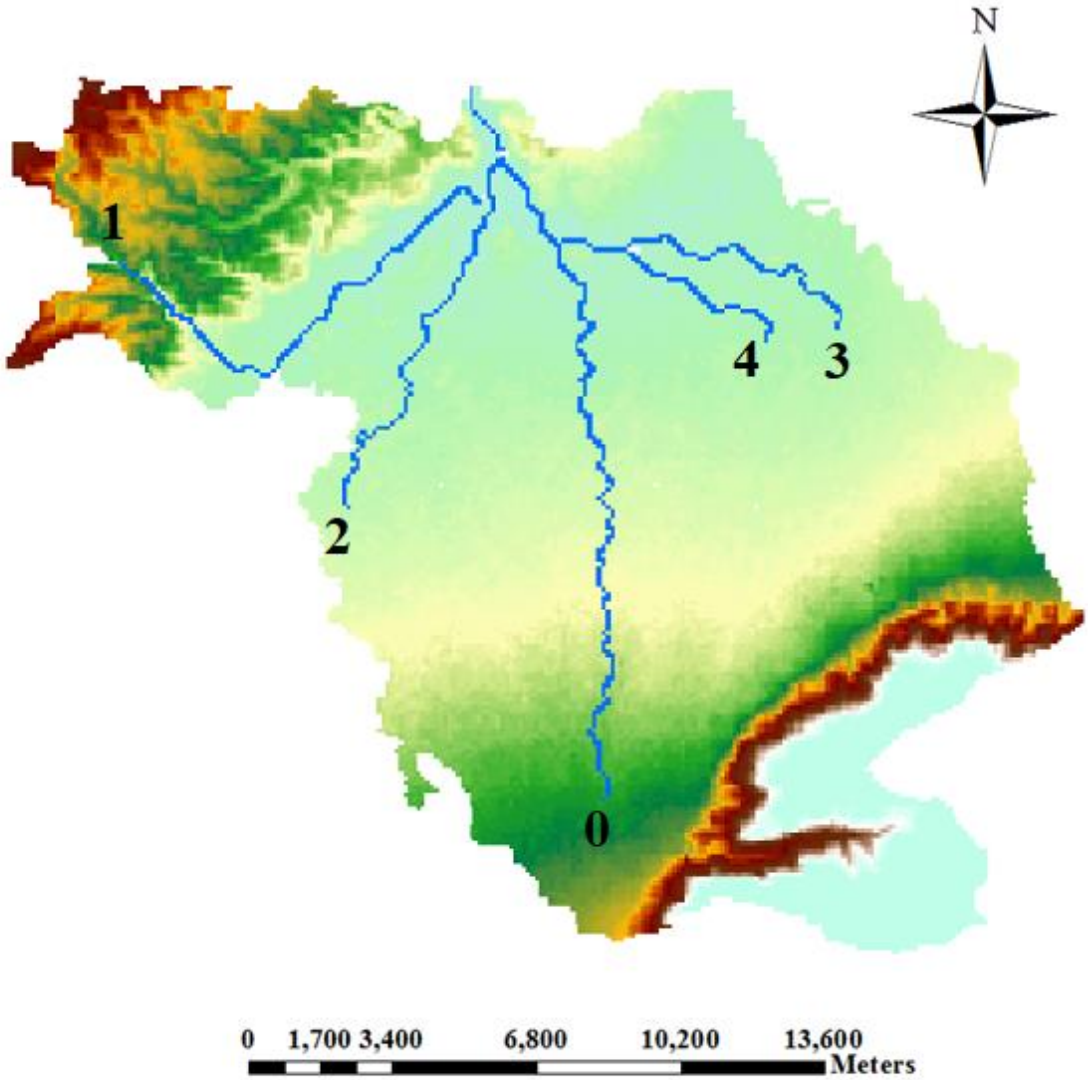


Fig.2.2 Watershed and river network.

2.2.3.2 Input data

All the data using in this study are the outcomes of evaluation study carried out in 2004-2011 by the cooperation team between ONEDD and JICA, on Harrach river pollution with various heavy metals caused by the industrial activities. The field sampling campaigns were conducted along Harrach River at different points according to the program established by ONEDD and the experts of JICA. Pb, Hg, Cr and Zn were considered for the simulation in study area. **Table 2.1** shows the major input sources of selected elements concentrations and flow rate (Q) data of wastewater discharged from factories at different points along the river network.

In simulation Case 1, the model was run with particular input data of, Pb, Hg, Cr, and Zn concentrations in wastewater discharged from the factories located along the study area (**Table 2.1**). This input data which were used in a previous simulation carried out by Bouragba *et al.* (2017) [8] with the addition of Cr and Zn data. Moreover, six factories which were not included in the previous simulation by Bouragba *et al.* (2017) [8], were added as a new point sources data (**Fig 2.3** and **2.4**).

Table 2.1: Input data of the concentration of wastewater discharged from the factories.

Factory name	Discharge location	Flow rate Q (m^3/day)	Pb (mg/L)	Hg (µg/L)	Cr (mg/L)	Zn (mg/L)
ENMTP**	river 0	3	0.20	8.36	0	0.01
ENPC**	river 0	/	0.27	12*	0	0
Raff Alger	river 1	7	0.51	18*	0	0
EMB1	river 1	320	2.40*	21.21*	0	1.47
BAG	river 1	100	0.45	3.54	0	0.60
SOACHLORE	river 1	930	0	5720*	0	0
Est KEHRI**	river 1	50	0.23	7.28	0.96*	0
Tan Semmache	river 3	30	2.23*	11.04*	60.43*	0
Tan KEHRI**	river 3	120	0.27	8	0.54*	0
AGENORE	river 3	3000	0.34	17*	0.15	1.08
CATEL	river 3	12	0.94*	0	0	0.63
AVENTIS**	river 3	/	0.38	0	0	0.71
ENAP**	river 3	7	0	12.11*	0	0
ENPEC	river 3	150	37*	10.23*	0	0
Hydrotraitment	river 3	/	22*	0	0.68*	54*

* Concentration values which are over concentration standards of the general regulations for wastewater qualities in Algeria (APPENDIX I and II).

** The factories which were not included in input data in the previous simulation done by Bouragba *et al*. (2017) [8].

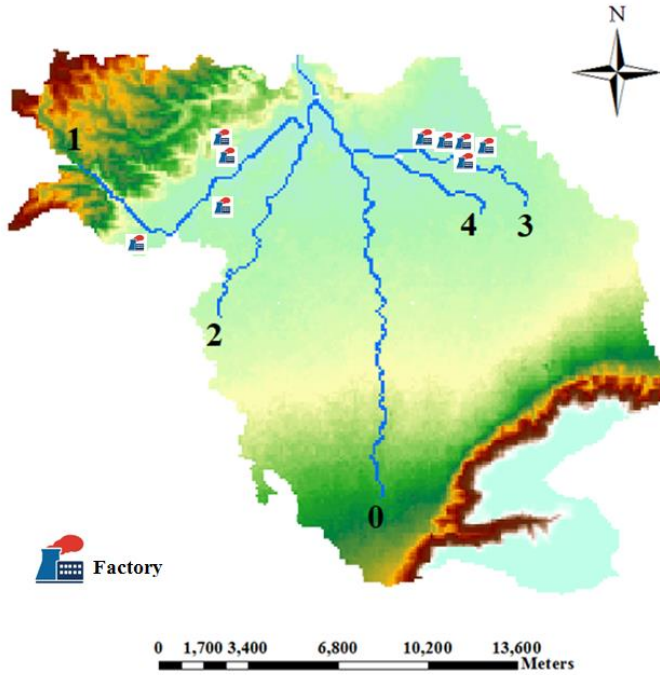


Fig.2.3 Distribution of major factories as point source of heavy metal pollution in the study area (factories included in previous simulation study carried out by Bouragba *et al.* 2017 [7].

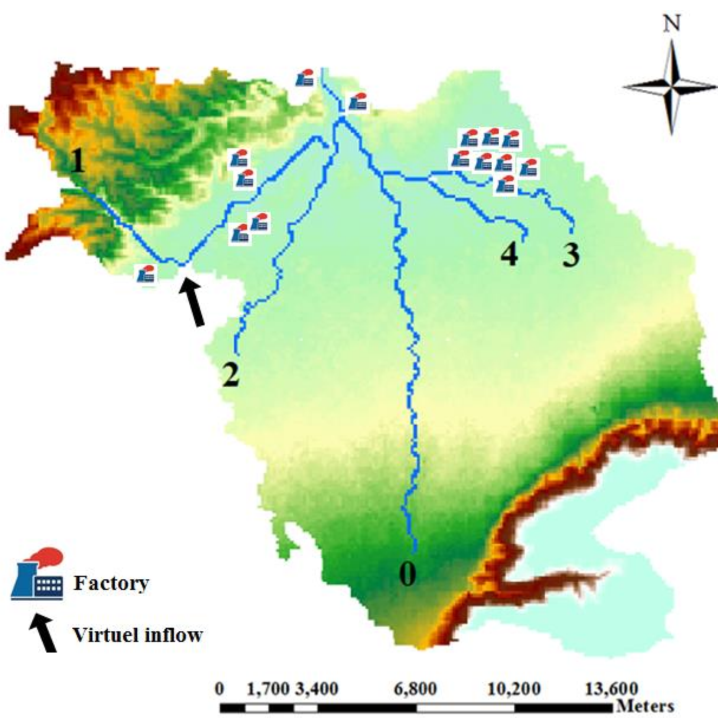


Fig.2.4 Distribution of major factories as point source of heavy metal pollution in the study area (factories included in present simulation study.

Table 2.2: Concentrations of each heavy metal in water at each observation point in study area.

	Location	Pb	Hg	Cr	Zn
		Water(mg/L)	*Water (mg/L)*	*Water (mg/L)*	*Water (mg/L)*
A1	river 0	0.73	24×10^{-4}	0.20	0.03
A2	river 0	0.60	18.2×10^{-4}	0.20	0.13
A3	river 0	0.20	10^{-3}	0.20	0.19
A4	river 0	0.89	12×10^{-4}	0.54	1.20
A5	river 1	0.20	10^{-3}	0.20	0.16
A6	river 1	0.57	72×10^{-4}	0.20	0.03
A7	river 3	0.89	10^{-3}	0.54*	0.20
EQS	/	0.50	5×10^{-4}	0.50	3

Table 2.3: Concentrations of each heavy metal in sediment at each observation point in study area.

	Location	Pb	Hg	Cr	Zn
		Sediment (mg/kg)	*Sediment (mg/kg)*	*Sediment (mg/kg)*	*Sediment (mg/kg)*
A1	river 0	200	3.40	640	1300
A2	river 0	83	0.50	168	218
A3	river 0	287	0.20	374	741
A4	river 0	170	0.30	190	1100
A5	river 1	144	0.80	68	211
A6	river 1	130	105	76	38
A7	river 3	142	0.20	521	741
EQS	/	218	0.71	370	410

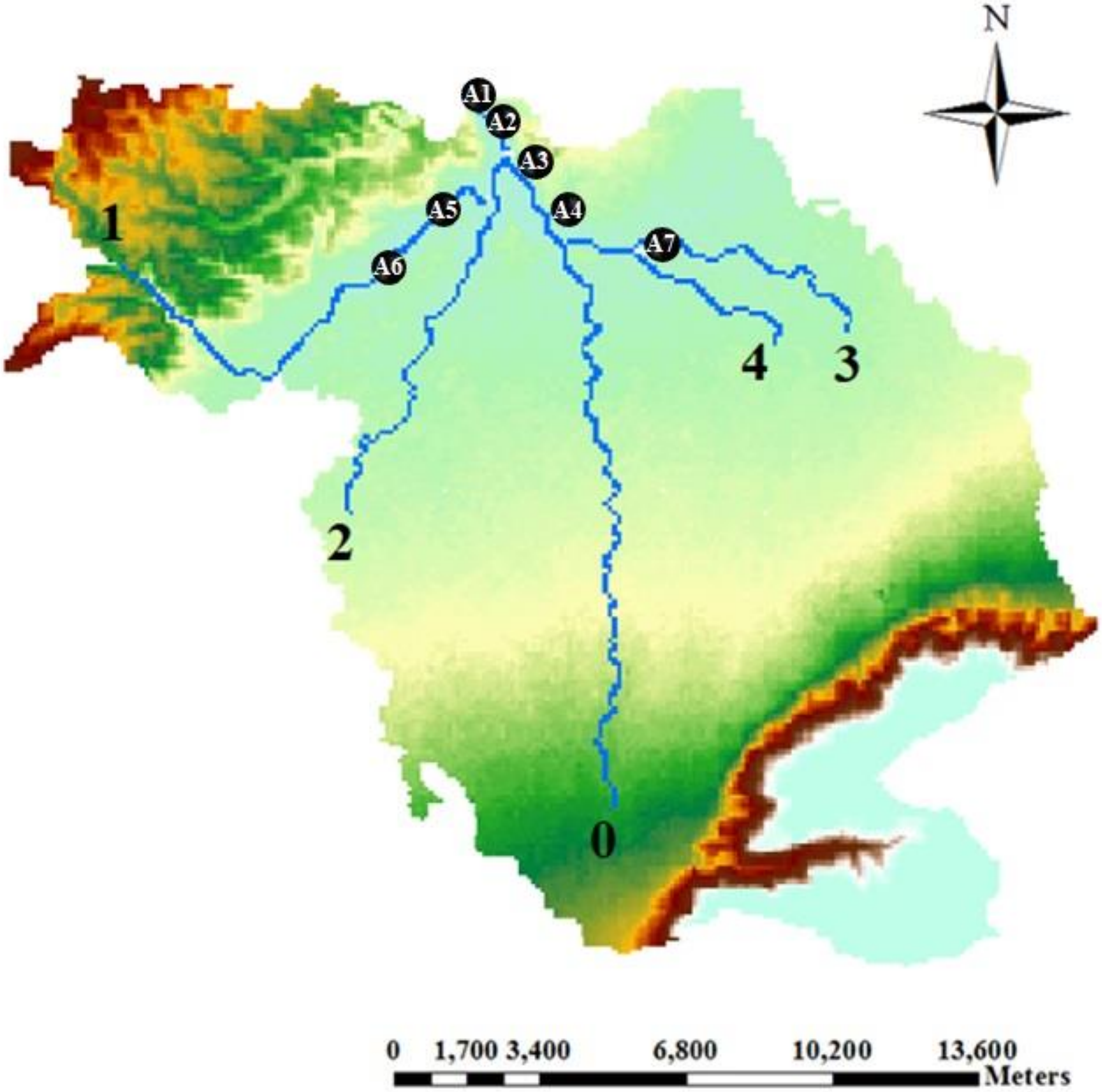


Fig.2.5 Observation points location in the study area.

2.2.4 Model set up

The simulation was carried out for the rainfall event of 2010 (ONM 2010). The rainfall data was given into the model with the interval of three hours. Based on effective width, the loss of water was estimated on the surface grid as 200 m. The routing of flood water was carried out on the surface grid that joins to the river grids and river flow occurs. The longwave equations are applied to estimate water level and discharge.

Pb, Hg, Cr, and Zn concentrations in water were modeled as instances of transportable objects. The interaction of metal concentrations between water and sediment was estimated using a partition model under the assumption of an equilibrium state.

Mathematica software has been used to analyze the process of each heavy metals transport in each river branch of study area.

In the upstream reach, there is a large catchment which is not industrialized or polluted. Therefore, the boundary inflow at the upstream end of river 1 was given a concentration of zero for all heavy metals. The discharge of 10^5 m^3/day was determined to be the same order of magnitude as the average river discharge (at the downstream end) multiplied by the ratio of the area of the upstream catchment to the area of the whole catchment, and was modeled as a virtual inflow at the river connection point of river 1, at the most-meandering point (**Fig.2.4**).

As shown in **Table 2.1** the discharge data were not available for some factories. These factories are located near to the rivers 0 and 3, where there are intensive other factories. Depending to the previous study [8], I assumed that the point source loads from these areas were 10^4 m^3/day at rivers 0 and 10^3 m^3/day at river 3, respectively, in order to represent the river discharge at the downstream end of river 0

The concentration of metals in sediment was estimated with a partition coefficient as described in equation **1.5** (chapter 1, sub. Section: heavy metal transport model):

The model was tested with average partition coefficient values estimated from the measurement data (source: ONEDD 2010) by using equation 1.4 (chapter 1) as K_{dPb}=430 (L/kg), K_{dHg}=520 (L/kg), K_{dCr}=950 (L/kg), and $K_{dZn}=4\times10^3$ (L/kg) for Pb, Hg, Cr, and Zn, respectively. However, in river 1, the concentrations in sediment were larger for Hg and lower for Cr compared with the concentrations in other rivers, (**Tables 2.2 and 2.3**). Also, the estimated K_d from the observation results shows at least two unrealistic fluctuations. Therefore, in order to ensure that results agree well with the verification data, the model was tested with average values $K_{dHg}=14\times10^3$ L/kg and $K_{dCr}=380$ L/kg for Hg and Cr, respectively, in river 1. These values were calculated from observation data in river 1.

Furthermore, a series of simulations cases were conducted in order to assess the effect of the uncertainty of the input point sources in the simulation and to assess the model performance in the simulation of multiple heavy metals with minimum input data. The model was run for seven different scenarios in following steps (**Table 2.4** and **Fig.2.6):**

- I Step 01: the simulation was conducted with adding new point sources (case 1) and without adding the new point sources (case 2).
- Step 02: the simulation was conducted with ignoring the pollution sources with high concentrations separately for each factory (cases 3, 4, 5, 6, 7),

It should be noted that the factories which were ignored in the simulation cases were chosen according to the magnitudes of the pollution they produced, i.e., AGENORE, SOACHLORE, Tan Semmache, and ENPEC have the largest source of Zn, Hg, Cr, and Pb, respectively. EMB1 has the second largest source of Zn and the third largest source of Pb and Hg.

- **Table 2.4:** A summary of different simulation cases.

Case	**Description**
Case 1	simulation of 4 elements (Pb, Hg, Cr, Zn) with the addition of 6 factories
Case 2	simulation of 4 elements (Pb, Hg, Cr, Zn) without adding 6 factories
Case 3	simulation of 4 elements (Pb, Hg, Cr, Zn) without AGENORE factory
Case 4	simulation of 4 elements (Pb, Hg, Cr, Zn) without EMB1 factory
Case 5	simulation of 4 elements (Pb, Hg, Cr, Zn) without SOACHLORE factory
Case 6	simulation of 4 elements (Pb, Hg, Cr, Zn) without Tan-Semmache factory
Case 7	simulation of 4 elements (Pb, Hg, Cr, Zn) without ENPEC factory

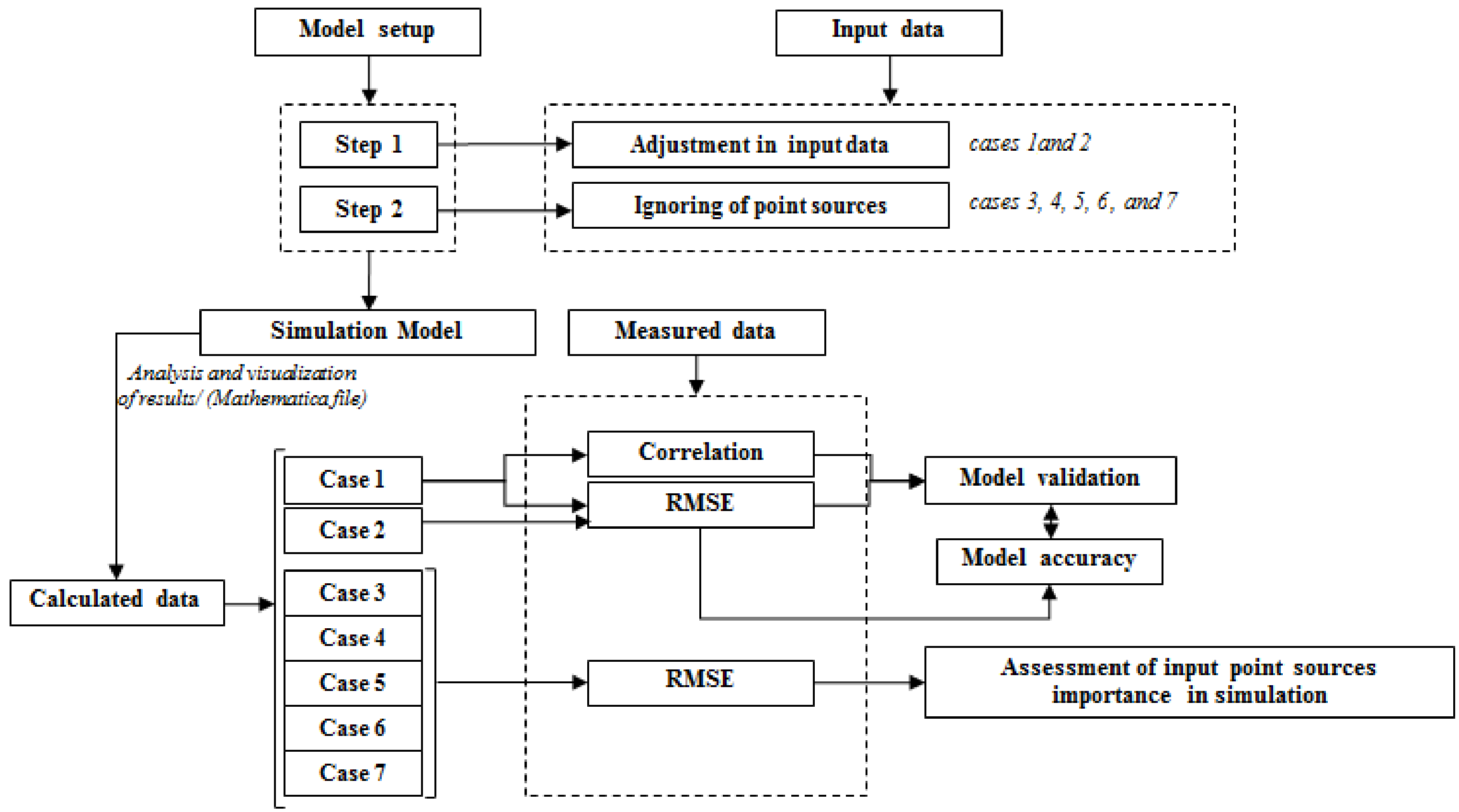


Fig.2.6 Simple chart for the simulation cases in this study.

2.3 Results and discussion

The transport of Pb, Hg, Cr and Zn concentrations originating from the industrial activities along Harrach River in the downstream direction were simulated. The simulation results of each metal concentration in water and sediment are shown in Figs **2.7** to Figs **2.10**, respectively, for case 1.

The calculated Root Mean Squared Errors (RMSEs) of metals concentration were lower than the EQS values for Pb, Cr, and Zn in both water and sediment, with values 9.7×10^{-2}, 5.2×10^{-2}, and 3.5×10^{-2} (mg/L) for Pb, Cr, and Zn, respectively, in water, and 74.85, 148.72, and 179.13 (mg/kg) for Pb, Cr, and Zn, respectively, in sediment. In contrast, the RMSEs values for Hg were higher than the EQS values in both stream water and sediment with RMSEs values i.e. 8.8×10^{-3} and 21.84, in water and sediment, respectively. (EQS values were shown in **Tables 2.2 and 2.3**).

The known point sources let to a large error in input data, which in turn may affected the simulation results, or the results were possibly affected by the greater uncertainties in the chemical behavior of Hg compared to the other elements (Pb, Cr, and Zn).

Figs 2.11 showed the correlations result between the simulation (case 1) and the measurements for the metal concentration in stream water. The correlations were high, i.e., 0.82, 0.95, and 0.98 for Pb, Cr, and Zn, respectively, otherwise it was lower for Hg, i.e., 0.35. In contrast, for sediment, the correlations results were more varied (**Figs 2.12)**, i.e., 0.25, 0.79, 0.53, and 0.67 for Pb, Hg, Cr, and Zn, respectively. This may be due to the effect of sediment properties and changes in physiochemical parameters on the metal's accumulation as well their concentration. Pb tends to be adsorbed to sediment organic matter more than to complex with organic matter in water. Furthermore, in constant pH, oxide and hydroxide of Pb have the larger solubility than those of Cr and Zn. The ion valence and sediment particle influence metal adsorption to the sediment particle, e.g. fine clay particle deposited in mild slope river can adsorb heavy metals more than sand particle [43].

In present simulation, Harrach River contamination with Pb, Hg, Cr and Zn was successfully simulated with only few observation data. Where the model flexibility of handing many transportable materials enhanced the model effectiveness simulate multiple heavy metals contaminations [8]. That is, column object which constitutes the calculation domain is generally incorporated in order to govern the vertical transport of physical quantities, along with a connection object which controls the horizontal transport of physical quantities between column objects [9]. Thus, various transportable quantities including the target chemical substances which are discharged in the watershed can be arbitrarily instantiated in column objects efficiently even when adding multiple metal elements.

Furthermore, the equilibrium assumption of chemical reactions in the calculation was suitable for predicting the metal transport in the river, because under the observed conditions, the interaction of heavy metal between water and suspended particle is sufficiently fast compared to metal transport.

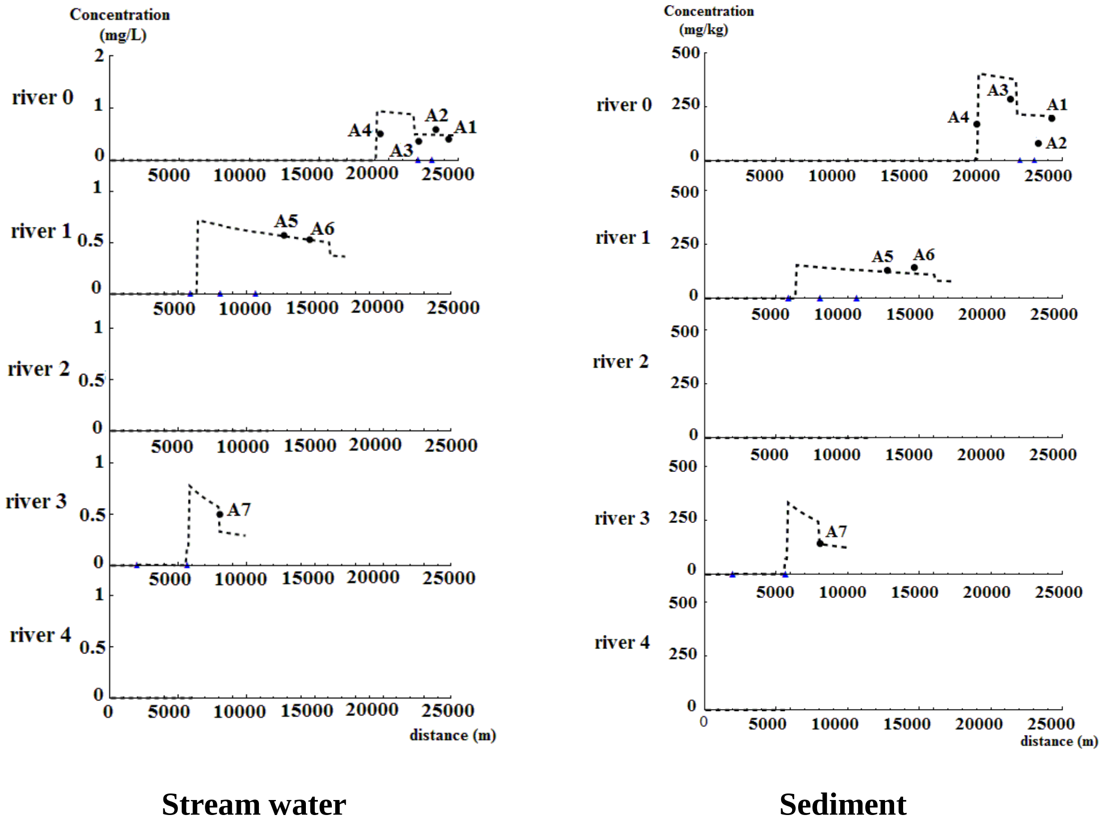


Fig.2.7 Simulation results for Pb concentrations in stream water (mg/L) and sediment (mg/kg) in the downstream direction; ▲: point source; ●: observation data; dashed line: simulation data (case 1).

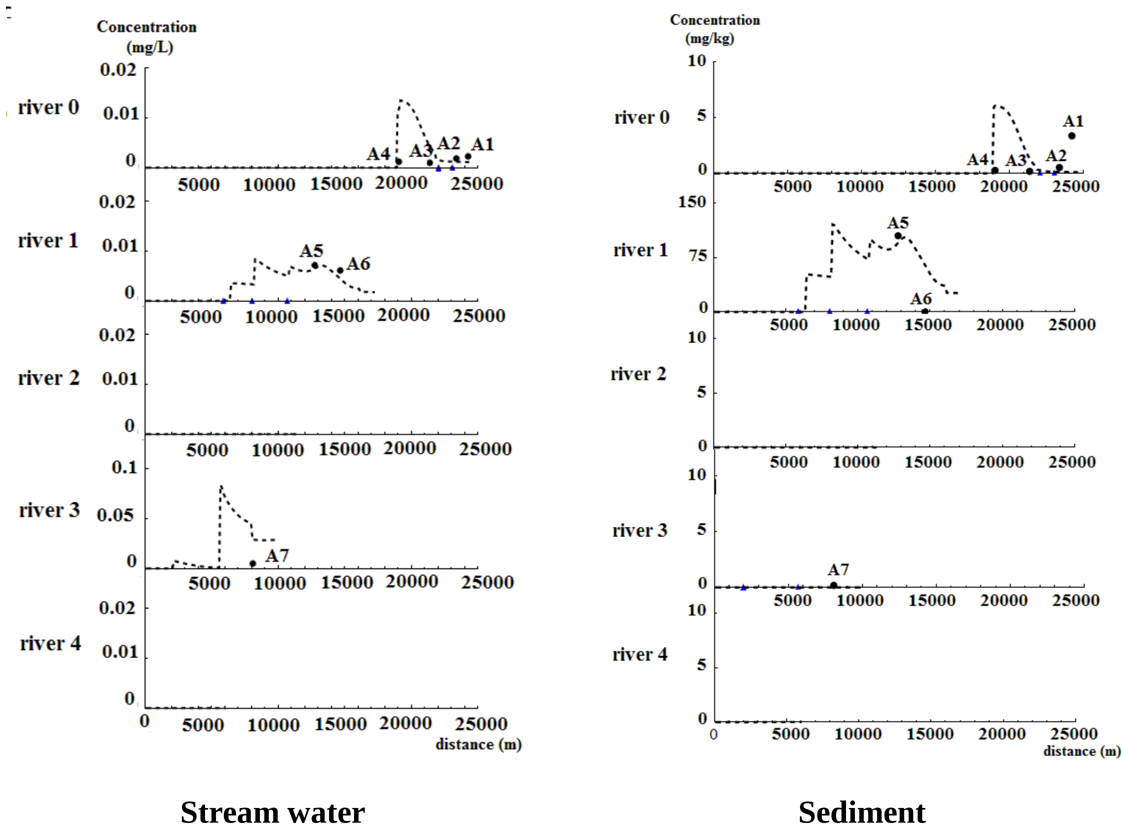


Fig.2.8 Simulation results for Hg concentrations in stream water (mg/L) and sediment (mg/kg) in the downstream direction; ▲: point source; ●: observation data; dashed line: simulation data (case 1).

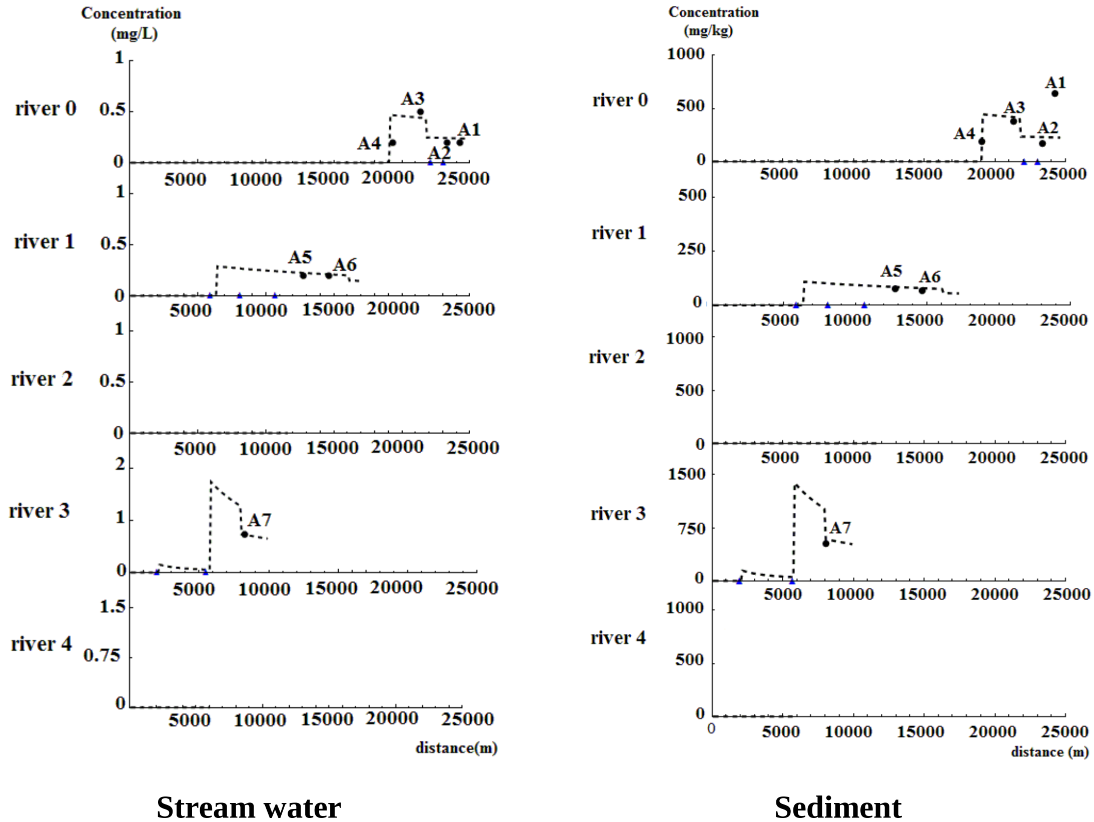


Fig.2.9 Simulation results for Cr concentrations in stream water (mg/L) and sediment (mg/kg) in the downstream direction; ▲: point source; ●: observation data; dashed line: simulation data (case 1).

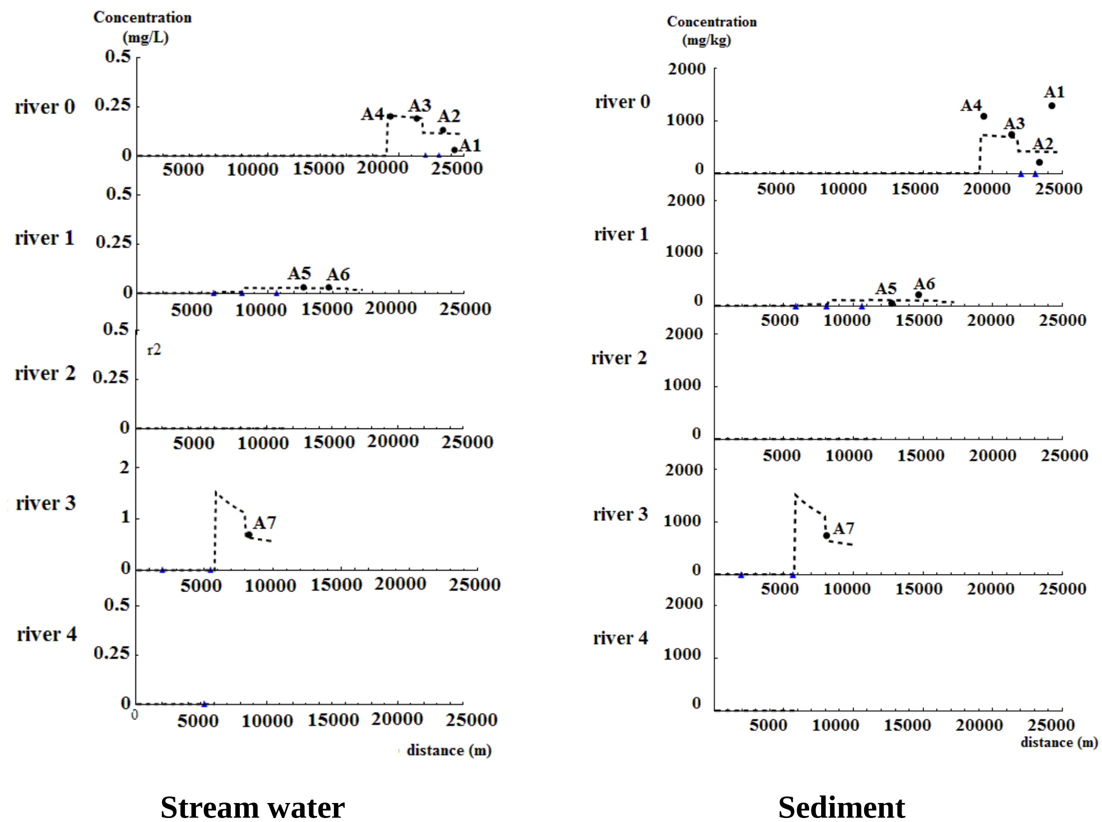


Fig.2.10 Simulation results for Zn concentration in stream water (mg/L) and sediment (mg/kg) in the downstream direction; ▲: point source; ●: observation data; dashed line: simulation data (case 1).

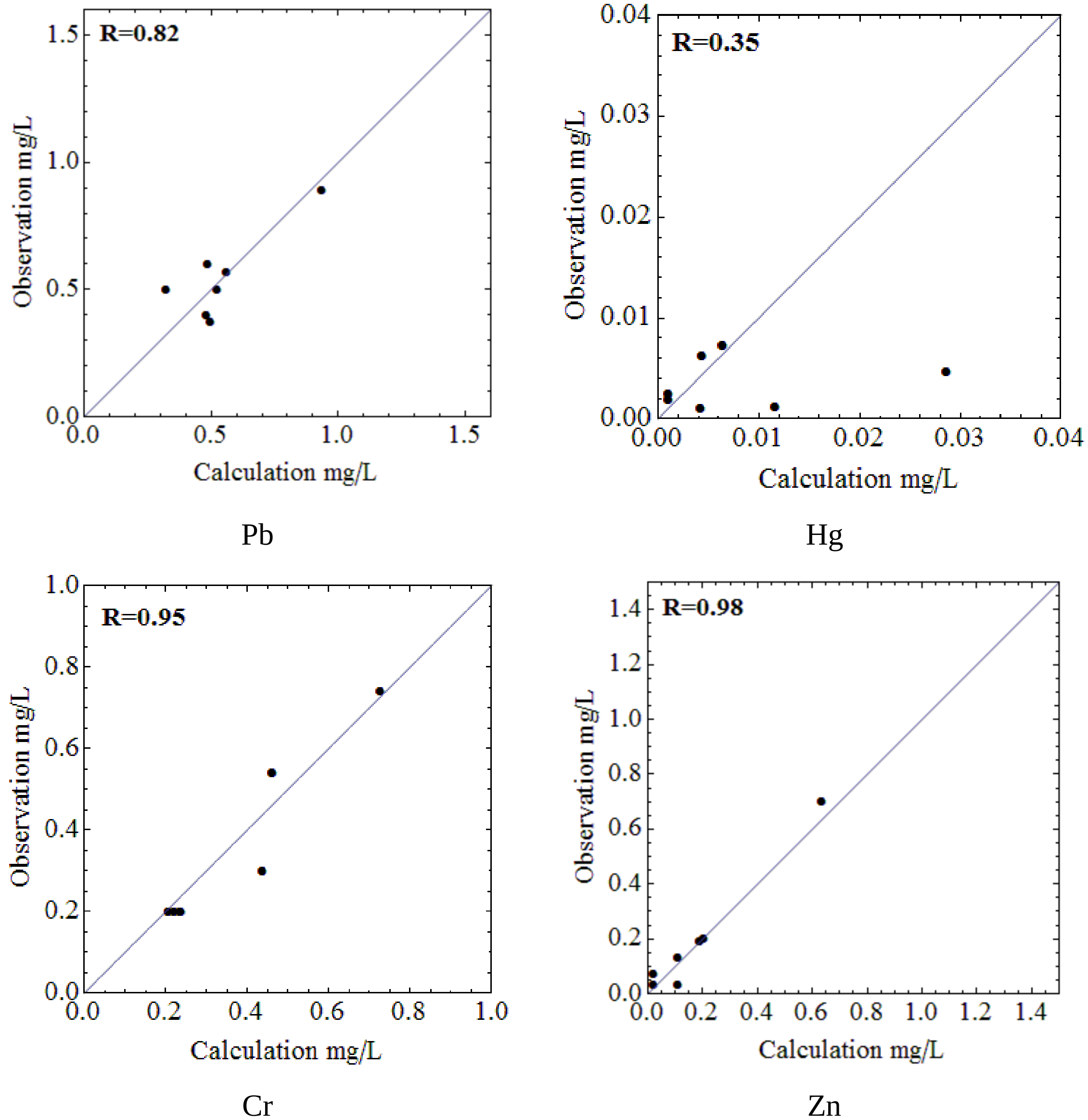


Fig.2.11 Correlation between simulation results (case 1) and observation results in each heavy metal concentrations in stream water (mg/L).

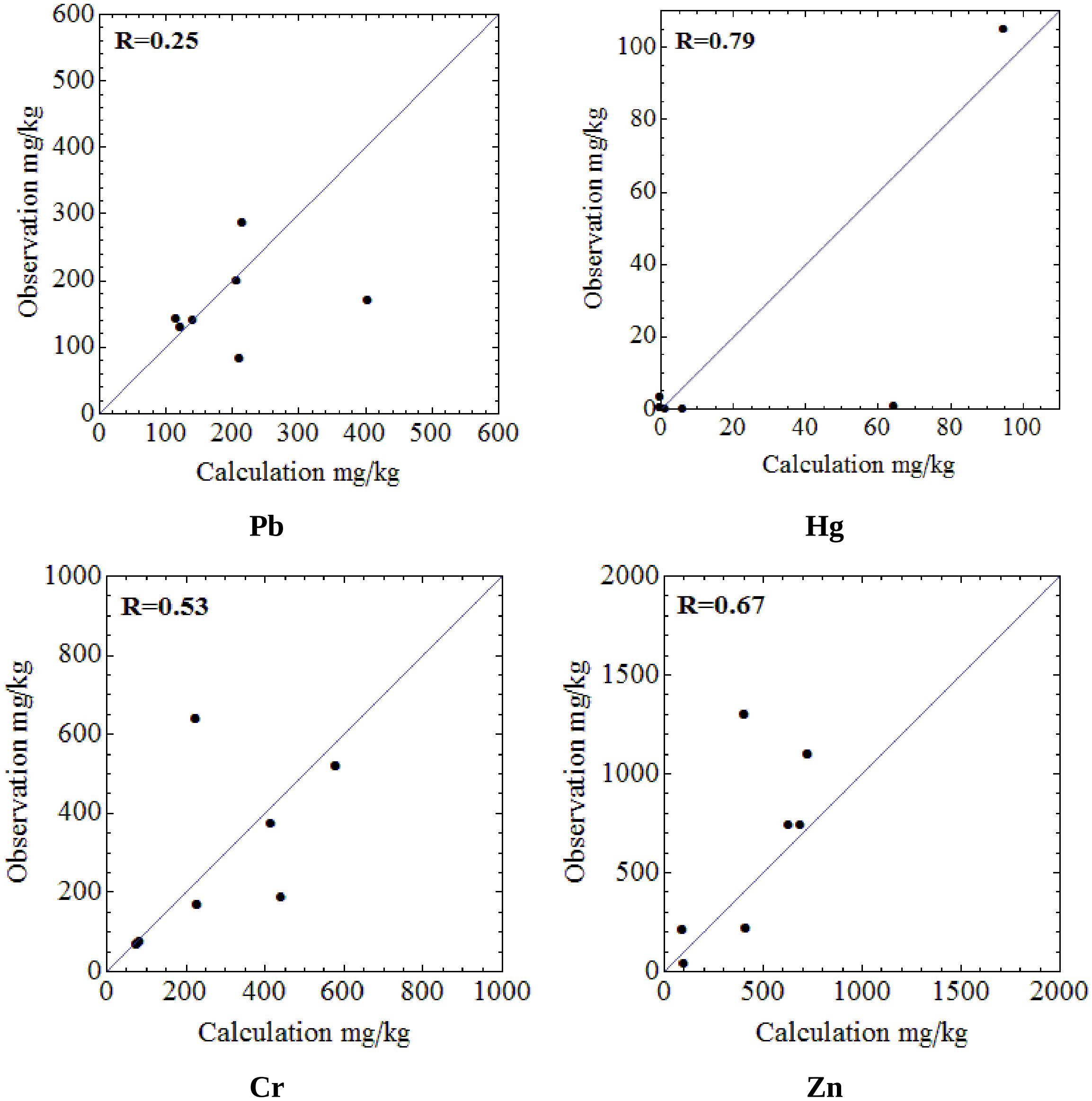


Fig.2.12 Correlation between simulations results (case 1) and observation results in each heavy metal concentrations in sediment (mg/kg).

Tables 2.5 and 2.6 showed the averaged RMSEs values for each simulation cases which were calculated using the original dataset at all the observation points in the study area in units of mg/L and mg/kg, in stream water and sediment, respectively.

Table 2.5: Calculated RMSE for each simulation case for each heavy metal concentration in stream water.

	Pb	**Hg**	**Cr**	**Zn**
	Water (mg/L)	Water (mg/L)	Water (mg/L)	Water (mg/L)
Case 1	9.7×10^{-2}	8.8×10^{-3}	5.2×10^{-2}	3.5×10^{-2}
Case 2	9.8×10^{-2}	8.8×10^{-3}	5.2×10^{-2}	3.5×10^{-2}
Case 3	9.8×10^{-2}	9.1×10^{-3}	5.2×10^{-2}	3.9×10^{-2}
Case 4	9.7×10^{-2}	9.3×10^{-3}	5.2×10^{-2}	3.6×10^{-2}
Case 5	9.8×10^{-2}	9.5×10^{-3}	5.2×10^{-2}	3.5×10^{-2}
Case 6	9.8×10^{-2}	9×10^{-3}	13.6×10^{-2}	3.5×10^{-2}
Case 7	11.1×10^{-2}	1.4×10^{-3}	5.2×10^{-2}	3.5×10^{-2}

Table 2.6: Calculated RMSE for each simulation case for each heavy metal concentration in sediment.

	Pb	**Hg**	**Cr**	**Zn**
	Sediment (mg/kg)	sediment (mg/kg)	sediment (mg/kg)	sediment (mg/kg)
Case 1	74.85	21.84	148.72	179.13
Case 2	74.89	21.86	148.79	179.32
Case 3	74.82	21.94	148.52	217.32
Case 4	74.94	13.43	148.66	200.23
Case 5	74.80	21.84	148.54	197.10
Case 6	74.89	22.37	153.73	197.36
Case 7	77.70	22.25	148.54	197.54

In simulation cases 1, The RMSEs were 9.7×10^{-3}, 8.8×10^{-3}, 5.2×10^{-2} and 3.5×10^{-2} for Pb, Hg, Cr, and Zn in water (mg/L), respectively, and 74.8, 21.8, 148.7 and 179.1, 179.3 for Pb, Hg, Cr, and Zn in sediment (mg/kg), respectively. In simulation cases 2, The RMSEs were

9.8×10^{-3}, 8.8×10^{-3}, 5.2×10^{-2} and 3.5×10^{-2} for Pb, Hg, Cr, and Zn in water (mg/L), respectively, and 74.9, 21.9, 148.8, and 179.3 for Pb, Hg, Cr, and Zn in sediment (mg/kg), respectively.

These RMSEs values show that the results of case 1 are slightly more accurate than for case 2, thus the additional six factories have not significantly an effect on the model performance.

The point sources with a high concentration and large discharge magnitude have a clear effect on the model accuracy, according to the simulation cases 3, 4, 5, 6, and 7. for both water and sediment results (**Tables 2.5 and 2.6)**.

For Pb, in simulation case 7, the RMSEs was the highest i.e., 11.1×10^{-2} (mg/L) and 77.70 (mg/kg) in water and in sediment, respectively. for the simulation results for cases 3, 4, 5, and 6 were slightly high. This is because in case 7, the ENPEC factory which is the highest source of Pb concentration, was omitted in calculation.

For Hg, the RMSEs results were more varied, especially in cases 4 and 7. ignoring the EMB1 in case 4 and ENPEC factory in case 7, decreased the RMSEs values, i.e., 1.4×10^{-3} (mg/L) in case 4 and 13.43 (mg/kg) in case 7, (Tables 2.5 and 2.6). In these RMSE results might be strongly affected by the large errors at the observation data which are located in rivers 1 and 3, since EMB1 is located in river 1 and ENPEC is located in river 3. Also, as we mentioned before there is an inconsistency between the partition coefficient of Hg in river 1 and the other rivers which may also be related to these errors. Based on these results, the errors in the input data for the point sources in river 1, and the errors in the observed data in water, and uncertainties in the chemical behavior of Hg, may be estimated.

For Cr, the Tan Semmache factory (this factory had the highest concentration of Cr among the other factories) has been omitted in case 6. The RMSEs values in this case were the highest, i.e., 13.6×10^{-2} (mg/L) in water and 153.73 (mg/kg) in sediment, respectively. The Cr discharged by the Tan Semmache factory into river 3 has a large impact for determining the distribution of Cr concentrations in the study area.

For Zn, the highest RMSEs was shown in case 5, i.e., 3.9×10^{-2} (mg/L) and 217.32 (mg/kg) in water and in sediment, respectively, where the EMB1 factory (which had the highest concentration of Zn among all factories) is omitted in this simulation case. The wastewater discharged by the EMB1 factory into river 1 is thus the most important for determining the Zn distribution of Zn concentration in the study area.

As showed above, omitting of high point source in simulation can compromise the performance of the model. Also, the simulation results may be affected by error caused by the quality of input data and/or verification data, as well the chemical behavior of each element such as non-equilibrium reactions, accumulation in sediment, etc. Mixing more than one Branch River can also lead to large variations in the results and affect the verification of the concentration of pollutants.

In the present study, the relation between different factories and the concentrations of Pb, Cr, and Zn in water and sediment was shown. However, the RMSEs for the simulation results evaluated only by the verification of dataset which contained irrelevant measurement points. For efficient and reasonable countermeasure of this, we need a densely monitoring of the concentrations in river 0, the middle reach of river 1, and the upstream reach of river 3, where large variations in the downstream direction were found, as shown in **Figs 2.7** to 2**.10**. Furthermore, although the simulations were shown to accurately capture the Pb, Cr, and Zn concentrations, the simulation of the Hg concentration recommended to be more carefully monitored.

Although the model was able to predict accurately the concentrations of Pb, Cr, and Zn, the simulation of the Hg concentration requires further consideration. However, the observation data with certain reliability in multiple heavy metals can be combined to be utilized as many apparent calibrations in order to adjust some unknown parameters from unrevealed point sources data (e.g. concentration and discharge), partition coefficient, etc. This is one advantage of using a simulation approach for multiple heavy metals in river basins with large sources of uncertainty. Also, in the aspect of environmental management and monitoring, the observation data of multiple heavy metals can be effectively used in model calibration for the future planning.

The simulation results obtained by Falconer & Lin (2003) [35] showed a good agreement with corresponding observed data, where they used salinity to model the partitioning coefficient of Cd and Zn in the Mersey estuary. The RMSEs values between the calculated results and measured data (i.e. 2.4×10^{-2} and 3.5×10^{-2} (mg/L) for Cd and Zn, respectively) are lower than corresponding EQS values (i.e. 0.2 mg/L and 3 mg/L for Cd and Zn, respectively). Also, Kashefipour & Roshanfekr (2012) [7] predicted the reaction coefficients variation for Pb and Cd in term of environmental factors by methodology based on the effect of environmental factors in the reaction coefficient in the advection-dispersion equation. In simulation results, the obtained The RMSEs were lower than EQS for both elements, i.e. 67.1×10^{-3} and 3.5×10^{-3} (mg/L) for Pb and Cd, respectively. Likewise, the obtained results for dissolved Pb, Cr and Zn in the present simulation (case 1) were good, with RMSEs values i.e. 9.7×10^{-2}, 5.2×10^{-2}, and 3.5×10^{-2} for Pb, Cr, and Zn, respectively.

In present simulation, the prediction of heavy metals concentrations in water and accumulated in sediment seems reasonable under the equilibrium assumption of chemical interactions of the heavy metals between the water and sediment. These interactions under observed conditions are faster than the transport of metal in the river. Therefore, the model could successfully simulate the concentration of heavy metals with greater accuracy than the EQS without including the dynamics of the chemical interactions.

The Geo-CIRC model in present study allowed the implementation of multiple elements (four heavy metals) although a few observation data are available. Whereas, the sufficient flexibility of the OOD to handle multiple elements as transportable instances, allowed the model to treat the selected heavy metals simultaneously but independently, these advantages cannot be achieved with other simulation system adopted with other studies (Falconer & Lin 2003; Kashefipour & Roshanfekr 2012) which are not OOD, and/or consider only single element with large requirement of large observation data among.

The systematic simulation cases adopted in the present study would help to provide a comprehensive description of the river contamination with various heavy metals which originated from industrial activities, although minimum amount of observation data available. It can thus provide results in a short time and reduce the number of samples which should be analyzed in situ. The application of the model in the monitoring methods can enhance and support environmental monitoring strategies for the contamination of rivers with heavy metals caused by industrial activities in developing countries where the observation data are limited.

2.4 Chapter Conclusion.

This chapter presents the detailed results of the application of Geo-CIRC model based on OOD for a numerical assessment of four heavy metals transport (Pb, Hg, Cr and Zn) in Harrach River. The model could successfully estimate the concentrations of Pb, Cr and Zn in both stream water and sediment of Harrach River in Algeria with a reasonable accuracy, whereas the simulation results for Hg concentration were lesser quality. The application of the model with systematic scenarios simulations allowed the implementation of an effective assessment methodology for data quality control and improving the monitoring practices. The results in present study proved the model effectiveness for monitoring of various heavy metal in rivers, that is because using of OOD with the Geo-CIRC supported the inclusion of multiple heavy metals in the simulation and improved the model flexibility even many unknown point sources exist.

Chapter 3

Environmental factors of partition coefficient for modeling of heavy metal concentrations in river sediment

This chapter is based on the results obtained by the published study "Empirical approach for modeling of partition coefficient on lead concentrations in riverine sediment", in International Journal of Environmental Science and Development, Vol 11 (7), pp: 352-357 . July 2020 doi: 10.181.78/ijesd.2020.11.7.1275.

3.1 Introduction

The sediment properties act as indicators of heavy metals pollution in aquatic system because a large part of the heavy metal input eventually accumulates in sediments [44]. In previous chapter 2, the concentrations of each heavy metals in sediment were estimated by using partition equation, however the calculated results were less quality. The use of constant K_d values in the calculation may related to the error in results. Depending on various environmental factors, K_d is not always constant, as well, affected by elements properties in the shape of solid and water phases [44]. According to Marco *et al.* (2000) [48], the particulate organic coating which represents 2% to 3% of the total SS are providing to the surface important characteristics in the exchange of trace metals between solid-water phases. As well, the SS in aquatic system have pH-dependent surface characteristics, that affecting the reaction of SS with certain functional organic contents groups. Also, the chemistry of organic contents surface results in the high pH-dependent adsorption [48].

Many studies related to the partitioning of heavy metals in aquatic system, demonstrated the influence of different physicochemical properties in the partitioning of heavy metals, and they were able to estimate the varying K_d values in term of different physicochemical properties such as pH, SS, etc.

For example, Rene *et al.* (1997) [45] have been determinate the K_d values of various heavy metals using multiple regression analysis, and they found that the determination of K_d values is strongly influenced by pH, whereas the organic carbon had also a distinct importance. Carlon *et al.* (2004) [46] have been predicted K_d values of Pb using regression equation with varied pH. Although they did not include additional variables except for pH, they believe that the application of chemometric methods to include the effect of OM on the partitioning of the metal in model seems promising.

In this chapter, I proposed a simple model of partition coefficient considering various physicochemical properties in riverine water and use it in simulation of Pb concentrations in sediment of Harrach River. That is, regression equations based on pH, SS, COD and BOD were derived to predict the varying K_d values, which in turn were incorporated in model calculation for Pb concentration in sediment.

3.2 Methods

In this Chapter, Pb was considered for calculation in the case study. The detail of the study area, model formulation, has been described in Chapter 2.

3.3.1 data preparation

Table 3.1 represents the data used for the verification of obtained results in this chapter. Whereas, the data showed in **Table 3.2** were used to calibrate and generate the appropriate regression equations between logK_d of Pb and the physicochemical properties such as pH, SS, COD and BOD, which are easily obtainable in common water monitoring.

Table 3.1: Pb concentrations in water and sediment, K_d and the physicochemical characteristics (pH, SS, BOD AND COD) (ONEDD 2010)

	Pb in water (mg/L)	Pb in sediment (mg/kg)	K_d (L/kg)	logK_d	pH	SS (mg/L)	BOD (mg/L)	COD (mg/L)
A1	0.4	200	500	2.4	7.6	100	45	690
A2	0.6	83	138	2.44	7.5	77	140	370
A3	0.37	287	776	2.14	7.4	170	39	220
A4	0.89	170	191	2.24	7.9	1500	420	140
A5	0.53	144	2712	2.6	7.7	190	22	0.5
A6	0.57	130	228	2.4	7.9	510	130	330
A7	0.8	142	178		7.2	1500	420	140

NB: these data are used for validation of regression equation, as well the verification of simulation results.

Table 3.2: Pb concentrations in water and sediment, K_d and the physicochemical characteristics (pH, SS, BOD and COD) (ONEDD 2006)

	Pb in water (mg/L)	Pb in sediment (mg/kg)	Kd (L/kg)	logK_d	pH	SS (mg/L)	BOD (mg/L)	COD (mg/L)
B1	0.6	137	228	2.4	7.48	18	110	15
B2	0.73	200	273	2.44	7.41	77	140	2825
B3	0.6	83	138	2.14	7.46	14	140	370
B4	0.54	86	159	2.24	8.04	77	56	130
B5	0.6	217	361	2.6	7.2	480	170	5400
B6	0.57	130	228	2.4	7.7	370	130	170

NB: these data are used for calibration of regression equation.

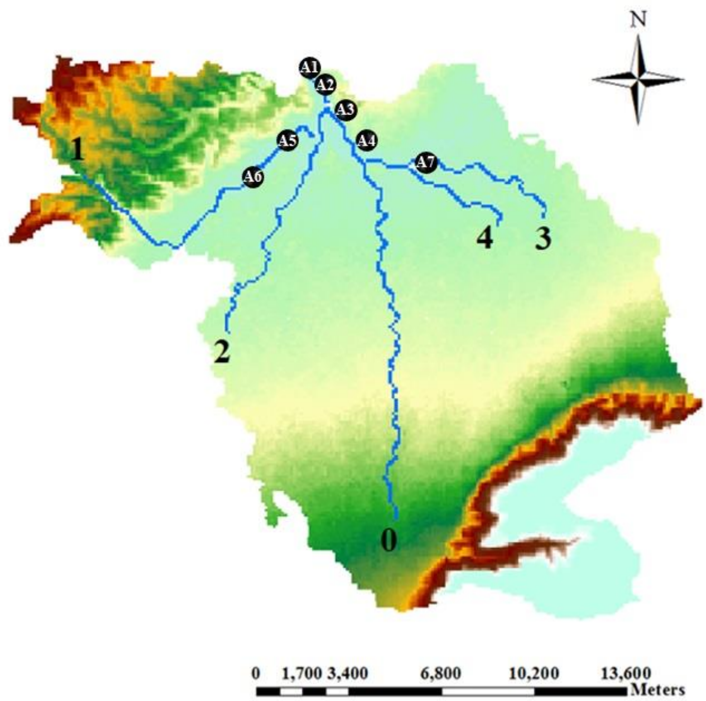


Fig.3.1 Observation point's location for data shown in **table 3.1**.

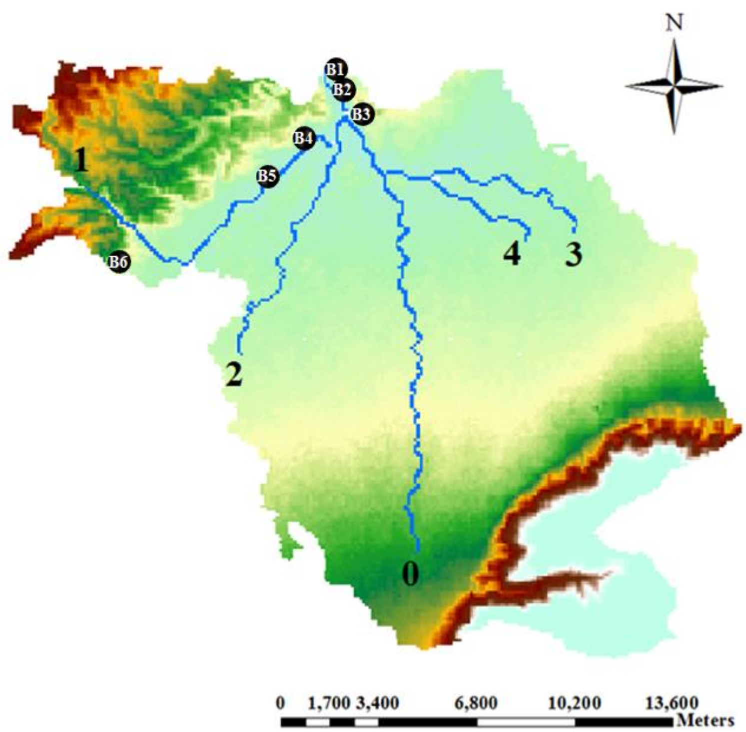


Fig.3.2 Observation point's location for data shown in **table 3.2**.

3.3.2 Multivariate regression model

In present approach, I have considered physicochemical parameters in the regression equation as explanatory variables. K_d values were calculated using the Equation **1.4** (given in chapter 1, sub. Section: heavy metal transport model).

The measured $\log K_d$ values were correlated to the physicochemical characteristics by multiple regression analysis by means of the Mathematica software. The coefficient of determination (R^2) is used to evaluate the linear relationship between calculated data and observed data. Furthermore, alternative regression models were separately generated in different cases, with ignoring one parameter for each case, in order to assess the influence of each parameter (pH, SS, COD and BOD) in the prediction of K_d values, as sensitivity analysis.

3.3.3 Simulation of Pb concentrations in sediment and model set up

This part of study focuses to improve the accuracy of Geo-CIRC for simulation of Pb concentrations in sediment, therefore the same procedures were performed in this part as in previous chapter (see Chapter 2, sections: model formulation and model set up) with considering the new calculation for K_d model. The transport of Pb concentration in water was modeled. Then, the concentrations of Pb in sediment were calculated using equation (**1.5**).

Two of simulation cases with K_d values for Pb were conducted.

- In the case 1, we used the average partition coefficient values K_{dPb}=430 (L/kg) in the same river basin.
- In case 2, we used the variable K_d values at each point location, those estimated from the empirical model in terms of pH, SS, COD and BOD. Here, observed physicochemical parameters were substituted in variable K_d model.

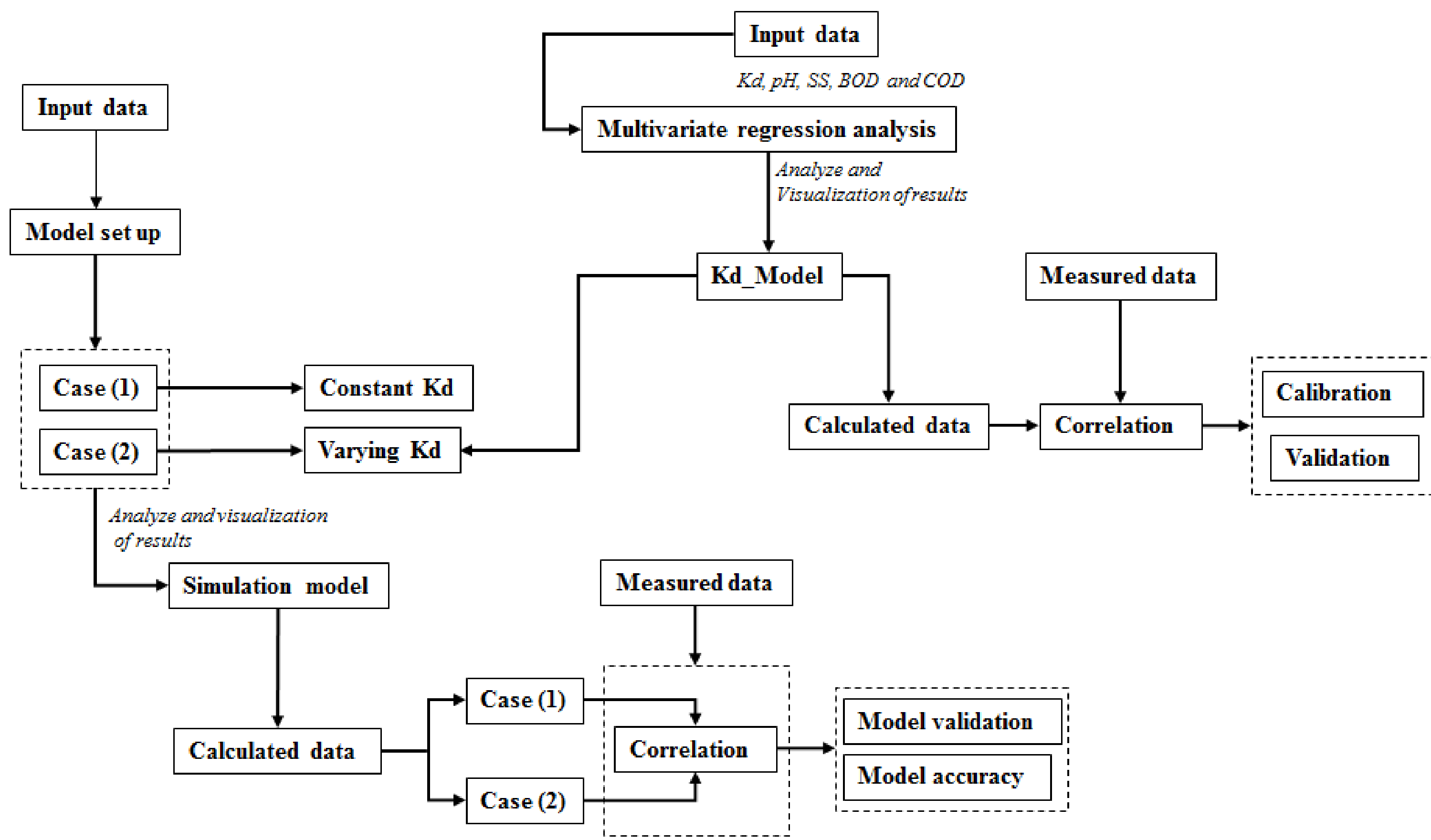


Fig 3.3 Simple chart for the simulation cases of Pb concentration in sediment.

3.3 Results and discussion

3.3.1 Multiple linear regression

In current study, the influence of pH, SS, and organic matter content on K_d of Pb were investigated. Thereupon, multivariate regression model of K_d (L/kg) were generated.

The comparison of sensitivities (regression coefficients) for each environmental factor in determination of $LogK_d$ values is shown in **Fig 3.4**. The results showed that the dependency of $logK_d$ has negative relationship with pH and BOD, whereas it has a positive relationship with SS and COD. The normalized regression values showed in **equation 3.3** indicated that the most influential factor in determining the partitioning of Pb between sediment and water is pH and BOD with regression coefficients of 0.165611 and 0.139133, respectively, following by SS with less influence, regression coefficient i.e. 0.101062, whereas COD has relatively no impact with regression coefficient 0.039254.

$$\log K_{d\ (Pb)} = 7.0013 - 0.139133BOD + 0.039254COD - 0.165611pH + 0.101062SS \qquad (3.3)$$

Where BOD, COD, and SS = the concentration of BOD (mg/L), COD (mg/L) and SS (mg/L), and pH = pH scale.

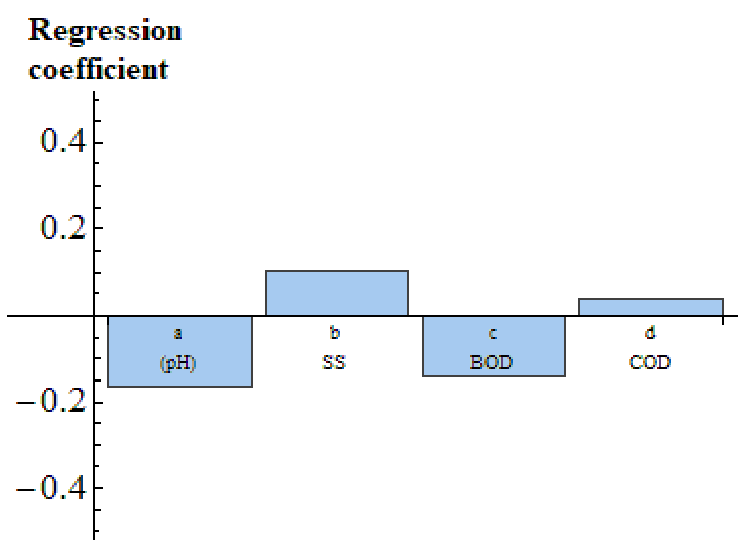


Fig 3.4 The comparison of sensitivities (regression coefficients) for each environmental factor in determination of $LogK_d$ values

The obtained regression equations for $\log K_d$ (L/kg) of Pb are shown in following:

$$\log K_{d\,(Pb)} = 7.0013 - 0.00359641BOD + 0.000178826COD - 0.572996pH + 0.000504306SS \quad (3.4)$$

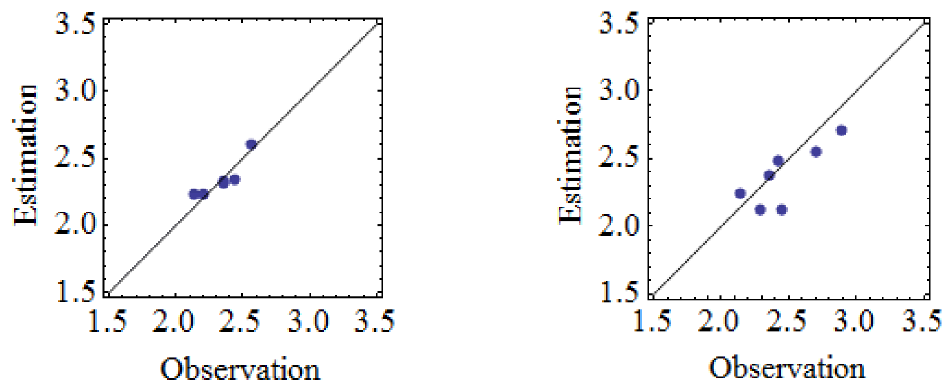
$$\log K_{d\,(Pb)} = 2.220920.000181906BOD + 0.0000413731COD - 0.000217953SS \quad (3.5)$$

$$\log K_{d\,(Pb)} = 2.91552 - 0.00126562BOD + 0.0000495889COD - 0.165611pH \quad (3.6)$$

$$\log K_{d\,(Pb)} = 3.11673 + 0.0000307791COD - 0.114511pH + 0.000257957SS \quad (3.7)$$

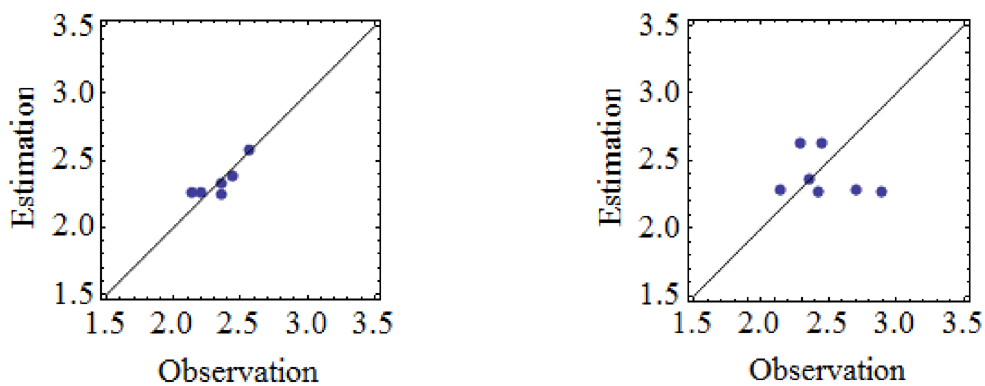
$$\log K_{d\,(Pb)} = 8.27116 - 0.00430179BOD - 0.728905pH + 0.00062743SS \quad (3.8)$$

Where BOD, COD, and SS = the concentration of BOD (mg/L), COD (mg/L) and SS (mg/L), and pH = pH scale.



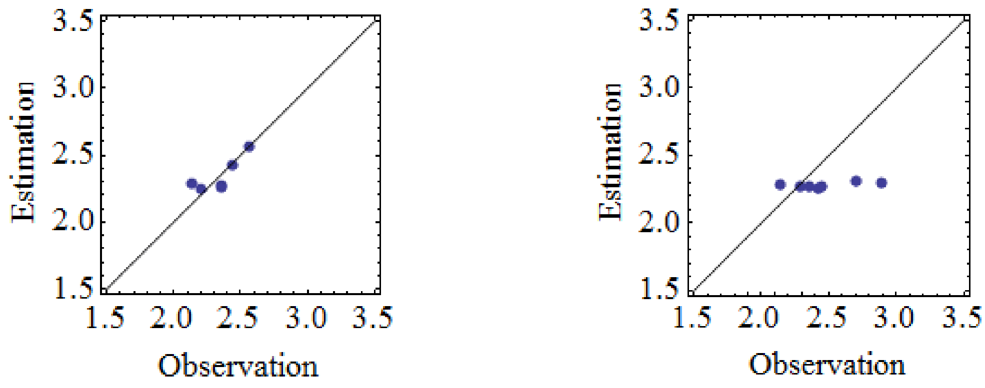
Calibration $R^2 = 0.80$ Validation $R^2 = 0.61$

Fig 3.5 Observed versus estimated $\log K_d$ values (under pH, SS, BOD and COD) which calculated according to the regression equation (**3.4**)



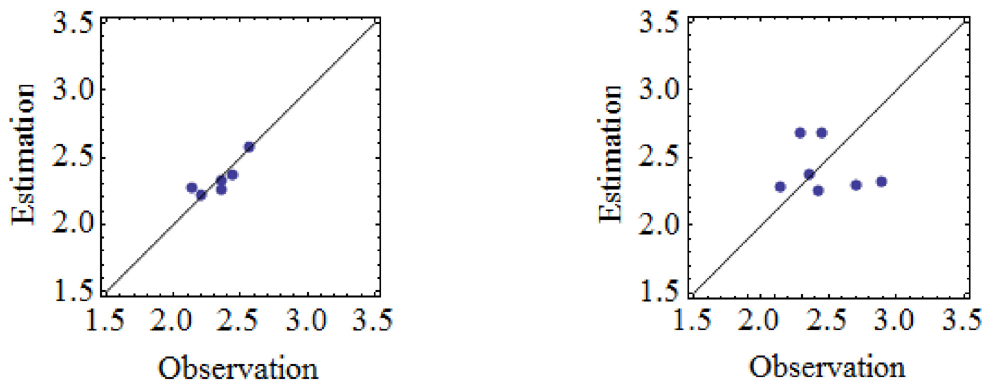
Calibration $R^2 = 0.70$ Validation $R^2 = 0.09$

Fig 3.6 Observed versus estimated $\log K_d$ values (under SS, BOD and COD) which calculated according to the regression equation (**3.5**)



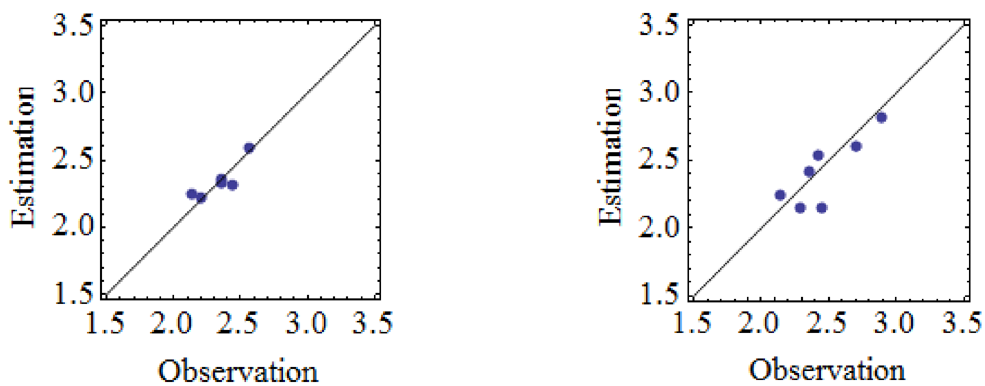
Calibration $R^2 = 0.65$ Validation $R^2 = 0.33$

Fig 3.7 Observed versus estimated logK_d values (under pH, BOD and COD) which calculated according to the regression equation (**3.6**)



Calibration $R^2 = 0.71$ Validation $R^2 = 0.06$

Fig 3.8 Observed versus estimated logK_d values (under pH, SS and COD) which calculated according to the regression equation (**3.7**)



Calibration $R^2 = 0.76$ Validation $R^2 = 0.67$

Fig 3.9 Observed versus estimated logK_d values (under pH, SS and BOD) which calculated according to the regression equation (**3.8**)

In calibration, the calculated K_d results were in good agreement with observation data given in **table 3.1** in equation **(3.4)** (R^2=0.80) when taking into account all selected parameters. In contrast, the results were slightly worse when we ignore one parameter in the calculation, with R^2.i.e. 0.70, 0.65, 0.71, and 0.76, when pH, SS, BOD and COD is not taken into account in each case, respectively (the left panels of **Figs 3.5** to **3.9**).

In validation, the results were shown in the right panels of **Figs 3.5** to **3.9**, where I used the data shown in **table 3.1**. The results showed a reasonable estimation of K_d values when taking into account all the selected physicochemical properties in equation **(3.4)** with R^2=0.61. In contrast, the results in other regression equations showed that COD was not significant parameter, where the results slightly improved rather than affected when diminishing COD (R^2=0.67) (**Figs.3.8**) comparing to pH, SS and BOD. That's maybe due to the possibility of the effect of inorganic contents to the COD values, since COD is the total measurement of all chemicals (which included organic and inorganic contents) in the water that can be oxidized [46]. The weak of inorganic content-dependency compared to organic content may led to decrease the importance of COD.

3.3.2 Simulation of heavy metals concentrations in sediment

Fig 3.10 shows the comparison between K_d distribution which used in both simulations cases 1 and 2 in the downstream direction of each river branch.

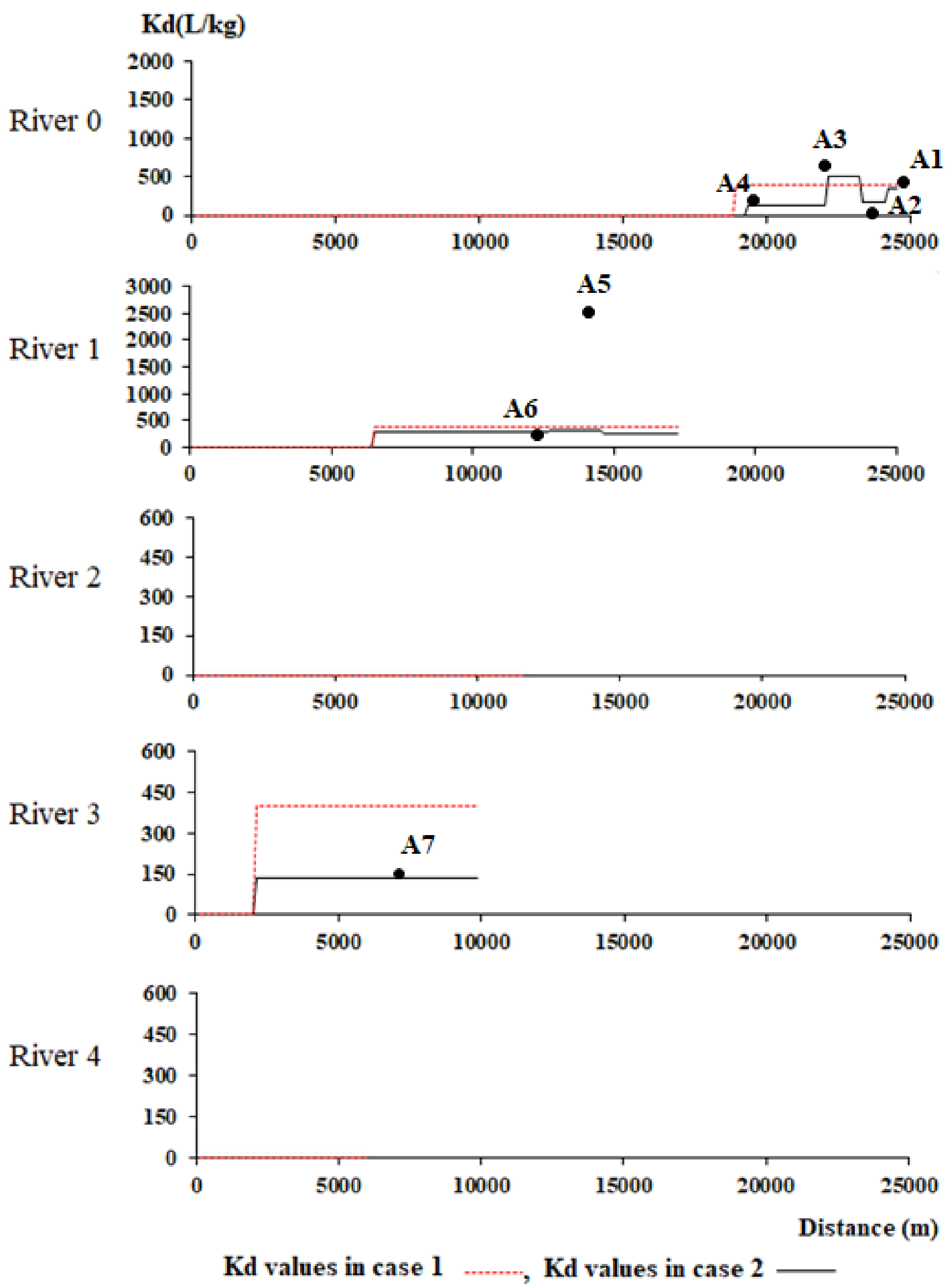


Fig 3.10 K_d distribution in the downstream direction of each river branch.

Fig 3.11 shows the simulation results for Pb concentrations in sediment, in the downstream direction. Pb concentrations of in sediment along Harrach River were predicted.

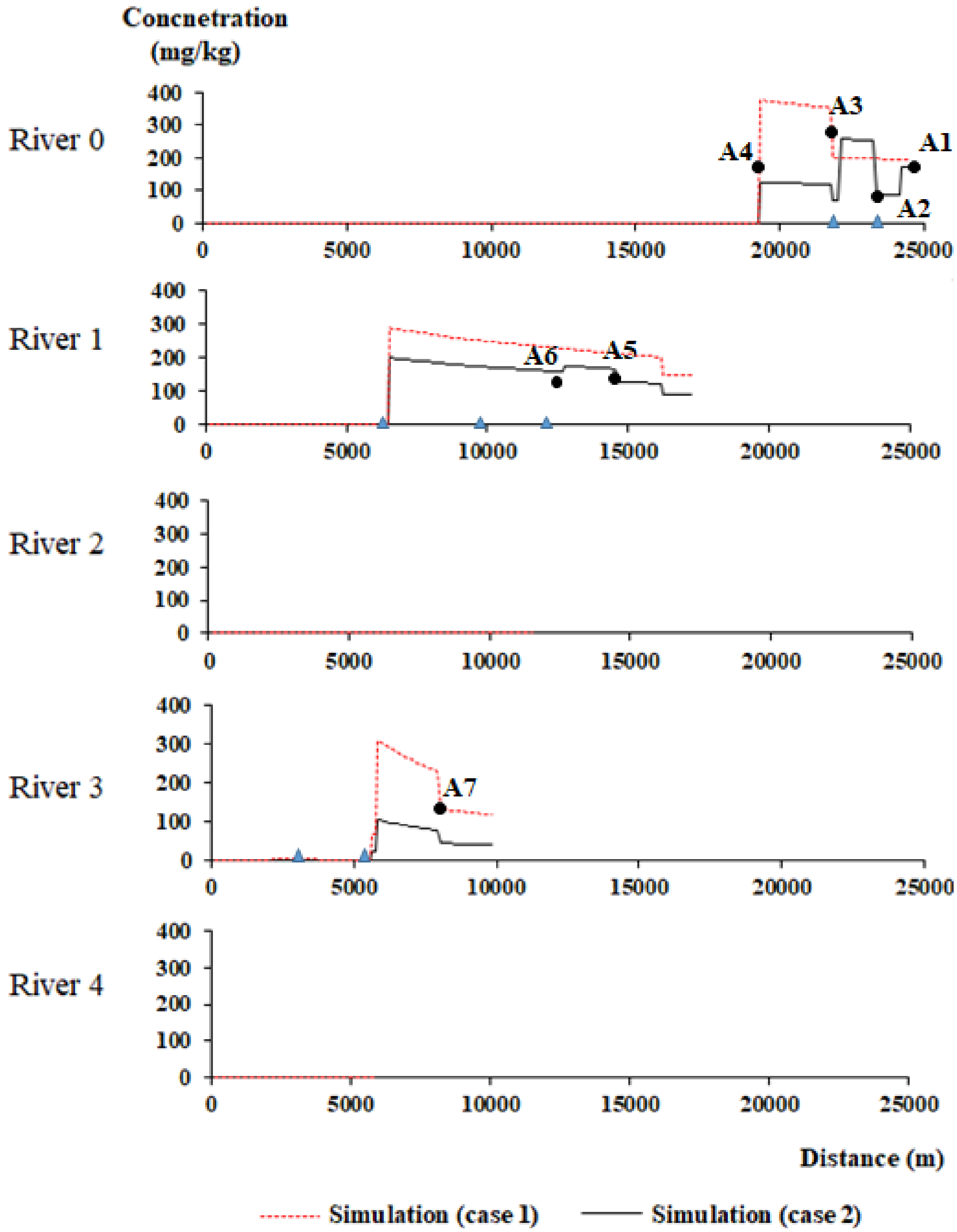
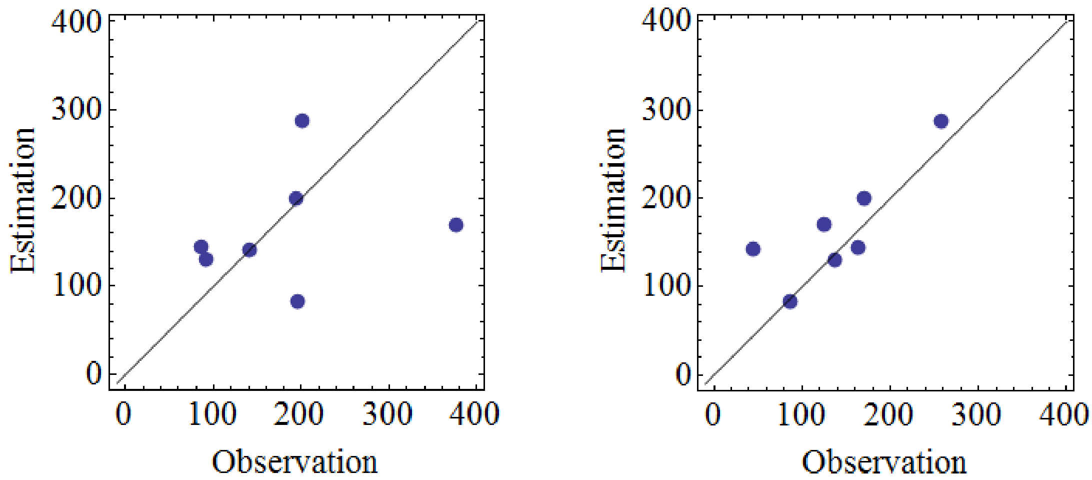


Fig 3.11 Pb concentrations pattern in sediment in the downstream direction of each river branch.



(Case 1) R^2 =0.05

(Case 2) R^2 =0.65

Fig 3.12 Observed versus simulated concentration (mg/kg) of Pb in sediment.

In the present simulation, the proposed approach was verified with measured data of Pb concentration, whereas the results of both cases were compared with the corresponding measured values given in **Table 3.1**.

The results in case 2 are more agreeing with corresponding measured data comparing to the results in case 1 where the model accuracy increased when we incorporated the varying K_d values those calculated using equation (6) i.e. R^2 0.05, and R^2 0.67, for case 1 and 2, respectively, as shown in **Fig 3.12**.

Using of constant K_d values that calculated from observation data in the proposed methodology in previous chapter 2 for predicting the concentration of heavy metal in sediment is relatively affected the model accuracy, because K_d value is not always constant, and it changes depending on the properties of elements and the characteristics of solid and water phases [44]. Therefore, the importance of the proposed K_d_model considering various environmental parameters (pH, SS, and OM content) is evident in its estimation of varied K_d values more accurately. Thereupon, the proposed K_d_model seems promising in heavy metals modeling field, where it can be helpful for more accurate simulation results of heavy metals concentration in river sediment.

Consequently, the simulation accuracy of Pb concentrations in sediment could be possibly improved by using K_d model calculated from BOD, COD, pH, and SS. However, it should be noted that there are limitations in using of this regression model for the present model scheme. That is, steady flow and uncomplicated distribution of physicochemical properties in rivers were required because physicochemical properties were not dynamically simulated in the present model scheme. In present model, observed physicochemical data were substituted into K_d model as model parameters. Fortunately, the assumption seemed suitable in Harrash River where the anthropogenic pollutant source might strongly influence, especially in dry season in semi-arid region.

3.4 Chapter conclusion

In this chapter, a simple approach for K_d model considering of various physicochemical properties (namely: pH, SS, BOD and COD) has been proposed, and used in simulation of Pb concentrations in sediments of Harrach River. The obtained regression models were successfully used to estimate the varied partition coefficient values of Pb with physicochemical properties changes, whereas, introducing of these varied values in simulation of Pb concentration in sediment increased the result accuracy of the model.

Chapter 4

Monitoring and Modelling of Heavy Metals Transport for Environmental Management of Rivers

4.1 Overview

The question which should be asked in this study is: "***Why Harrach River in Algeria was the target river basin? And what advantage would the water quality management in development country be given by using of numerical models (Geo-CIRC model) and proposed assessment methodology?***"

There is an obvious weakness of water quality management pertaining to urban rivers in developing countries; the water quality monitoring is not being consequently organized without clear perspective because low level of regulatory compliance and environmental ethics, and undisclosed information on pollutant source, wastewater discharge, water qualities, and even location of factories. This is especially true in regarding to the policy of environmental monitoring adopting by the government in Algeria. This weakness causes the appalling situation in the ecosystem, for example in Harrach River where the pollutants produced by high intensity of industrial activities including undisclosed point sources are drastically transported and distributed along the river according to evaluation study of ONEED/JICA [21]. So that, the pollution level in river system is hardly controlled and becomes very high at here and there.

To rectify the problem of pollutants distribution along the river system, an intensive monitoring of the emissions of industrial activities is required at first, but the followed policy of the environmental management in Algeria as mentioned before is only rely on the non-systematic monitoring which are time-consuming, labor-intensive, and high cost. Therefore, the monitoring with use of simulation approach providing a continuous access to results regardless of different conditions and data available is promising in the future water quality monitoring. In this chapter we will try to present the benefits of using the numerical models (Geo-CIRC model) in monitoring system in Algeria, in order to highlight the practical outcomes of this study as future monitoring methodology for environment management in urban river basin in developing countries.

4.2 Reasons of including Geo-CIRC model in the proposed assessment method

Although, many studies have been proved the effectiveness of different numerical models in the prediction of heavy metals transport in aquatic systems (e.g. Falconer *et al.* (2005) [6] and Kashefipour & Roshanfekr (2012) [7], Florimond *et al.*, (1998) [31], Mark L.*et al.* (2008) [32], etc.), the proposed assessment method with Geo-CIRC model is possibly useful in monitoring system and effective countermeasure comparing to conventional models for the following reasons:

① The first main reason of using Geo-CIRC model is that it could simulate multiple elements with minimum additional coding. This model feature allows us to be able to simultaneously utilize much information which could be taken from limited monitoring data. It means that the multiple elements simulation can detect undisclosed pollutant sources (as shown in Chapter 2) as if it is multiple water qualities sensor, and it can severely find inconsistency between monitoring results and disclosed pollutant sources. Furthermore, the partition coefficient model (shown in Chapter 3) can enforce the availability of sediment data. These advantages do not exist in the other non-OOD modeling system (**Fig.4.1**).

② Comparing with the others numerical models, the Geo-CIRC model can provide comprehensive assessment of the environmental state in the river system with less cost in short time by using less efforts because of the OOD's advantage mentioned above. For example, it can provide detail information about not only water quality distributions along river system but also existing potential of undisclosed pollution sources (as shown in Chapter 2). Through them, we can know important location to be monitored in river system and to reduce or increase the number of monitoring point for efficient environmental management (as shown in **Figs. 4.2 and Fig.4.3**).

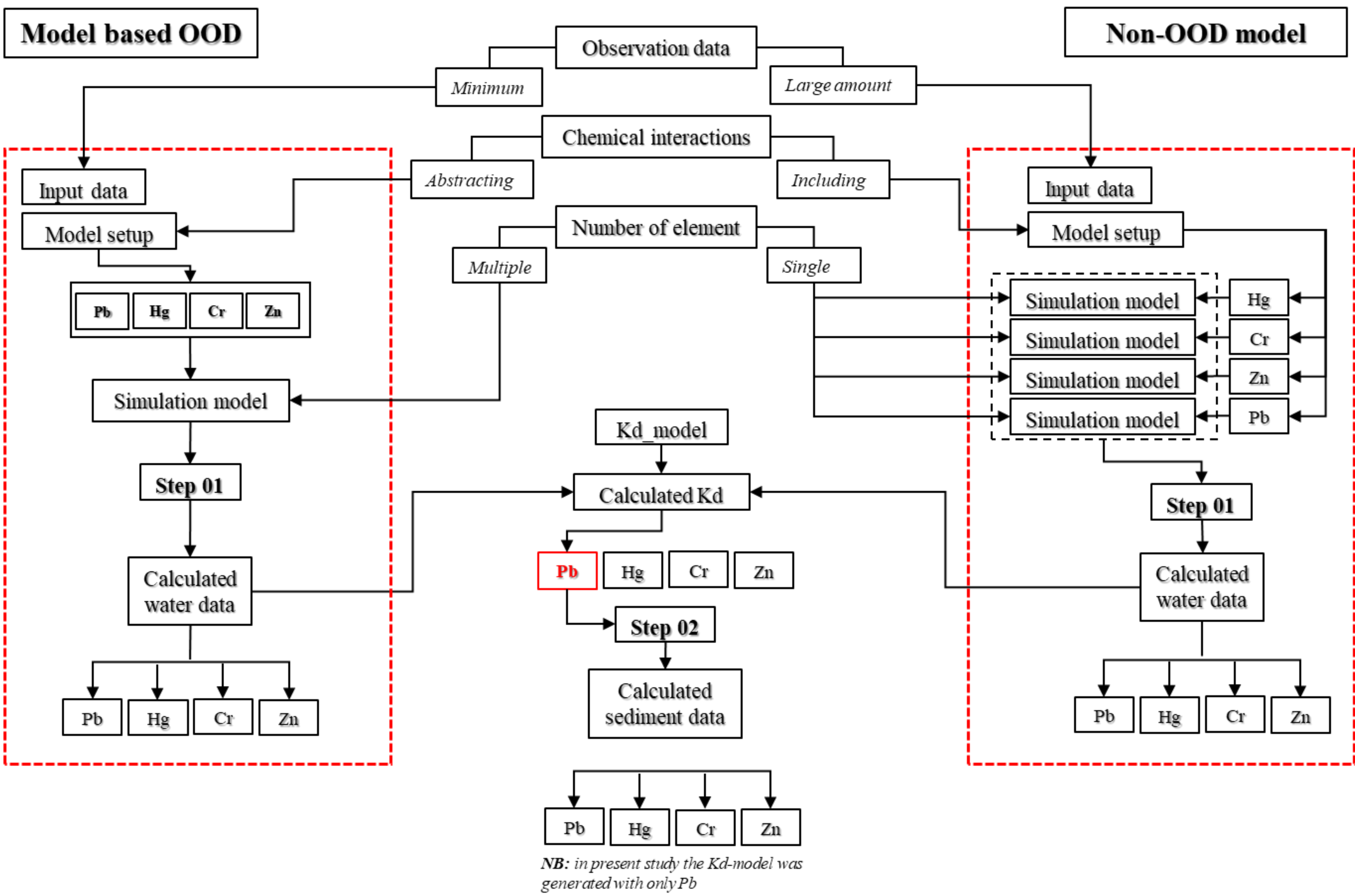


Fig.4.1 simple chart represents the comparison between Geo-CIRC model and conventional model.

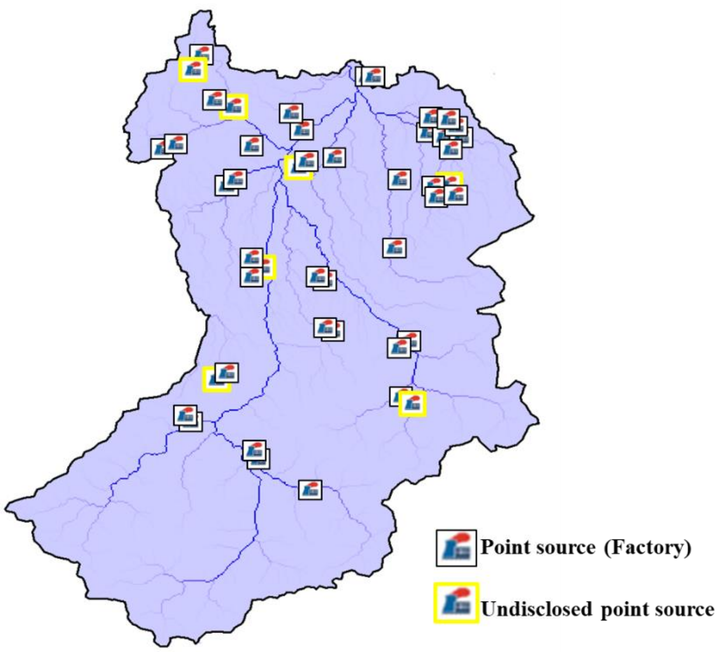


Fig. 4.2 virtual undisclosed point sources location.

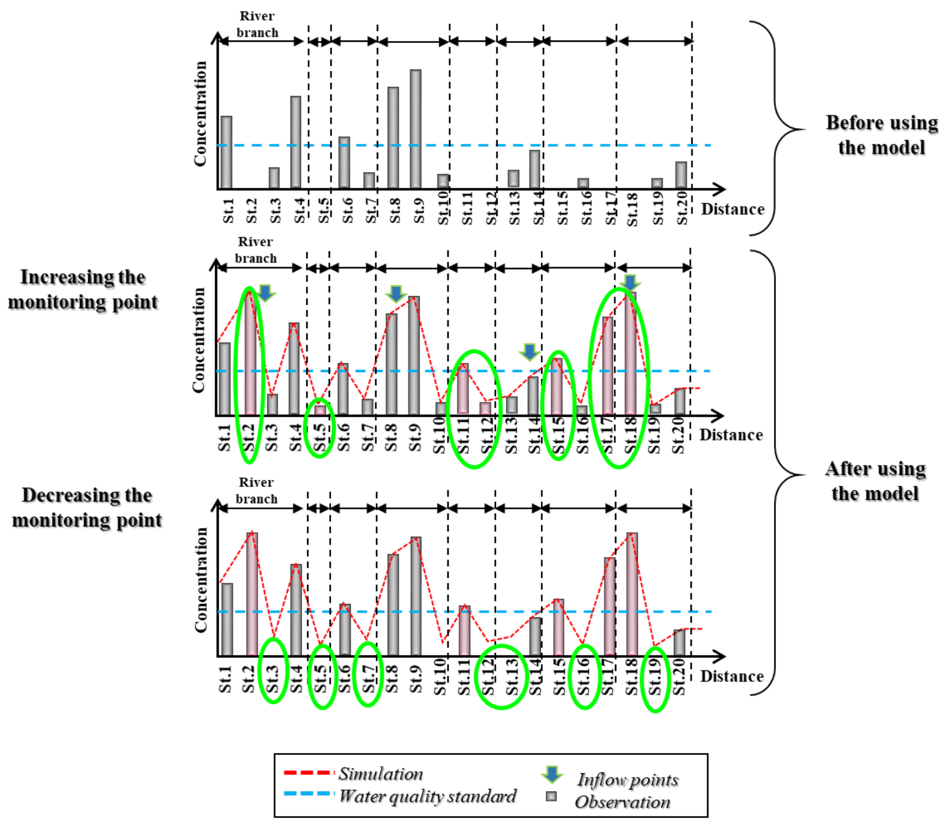


Fig. 4.3 Virtual graphs represent the increasing/decreasing of monitoring locations in site.

4.3 The benefits of using the Geo-CIRC model for the monitoring of rivers pollution in Algeria

Based on the outcomes obtained in present study, the including of the Geo-CIRC model in the proposed assessment methodology would having prominent benefits, amongst them:

① Better information with a new technological support for less cost and effort within a short time

The application of Geo-CIRC model in proposed assessment method in Harrach river case is the first work in its kind, as it never be used the modeling to assess the contamination of aquatic systems with heavy metals in Algeria before. Whereas, all the assessment studies were focused on observation data. Among these studies, we mention as examples, the assessment of heavy metals distribution in sediment of Algiers Bay in Algeria carried out by Atroune & Boutaleb (2012) [49]. Also, Louhi et al. (2012) [50], they determined some heavy metal in sediments in order to assess the pollution of Seybouse River in Annaba, Algeria.

The results obtained in this study would be more representative than ones obtained in the traditional way, and may allow to more detail representation of heavy metal contamination in the river basin with less cost in short time by using less efforts (**Fig.4.4**). Therefore, including of proposed simulation approach in monitoring system of heavy metals pollution in river basins would be an important starting point for reliable monitoring strategy, in order to confront the problem of river pollution with heavy metal caused by industrial activities in Algeria.

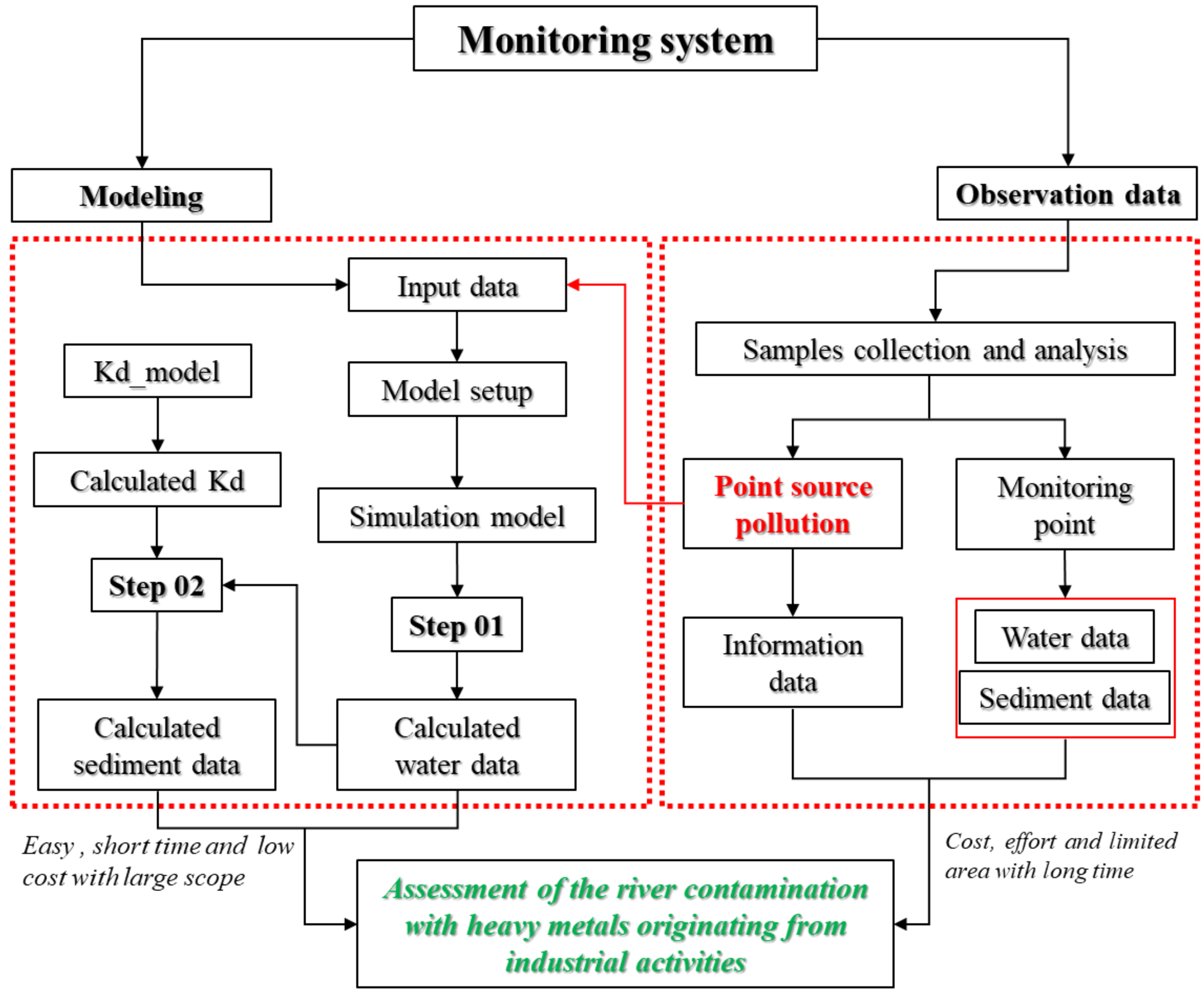


Fig.4.4 Simple chart for the assessment method using the simulation approach in present study.

② **Assess different type source contamination with heavy metals:**

The distributed type model like Geo-CIRC can predict large scale pollutant transport. It can provide a predictive description of not only from point source (e.g. industrial discharges impact) **(Fig. 4.5**) but also non-point source (e.g. air pollution, natural mineral) on the heavy metal levels in aquatic ecosystem. In addition, proposed method to evaluate the impact of pollutant source (shown in Chapter 2) is available both of point source and non-point source.

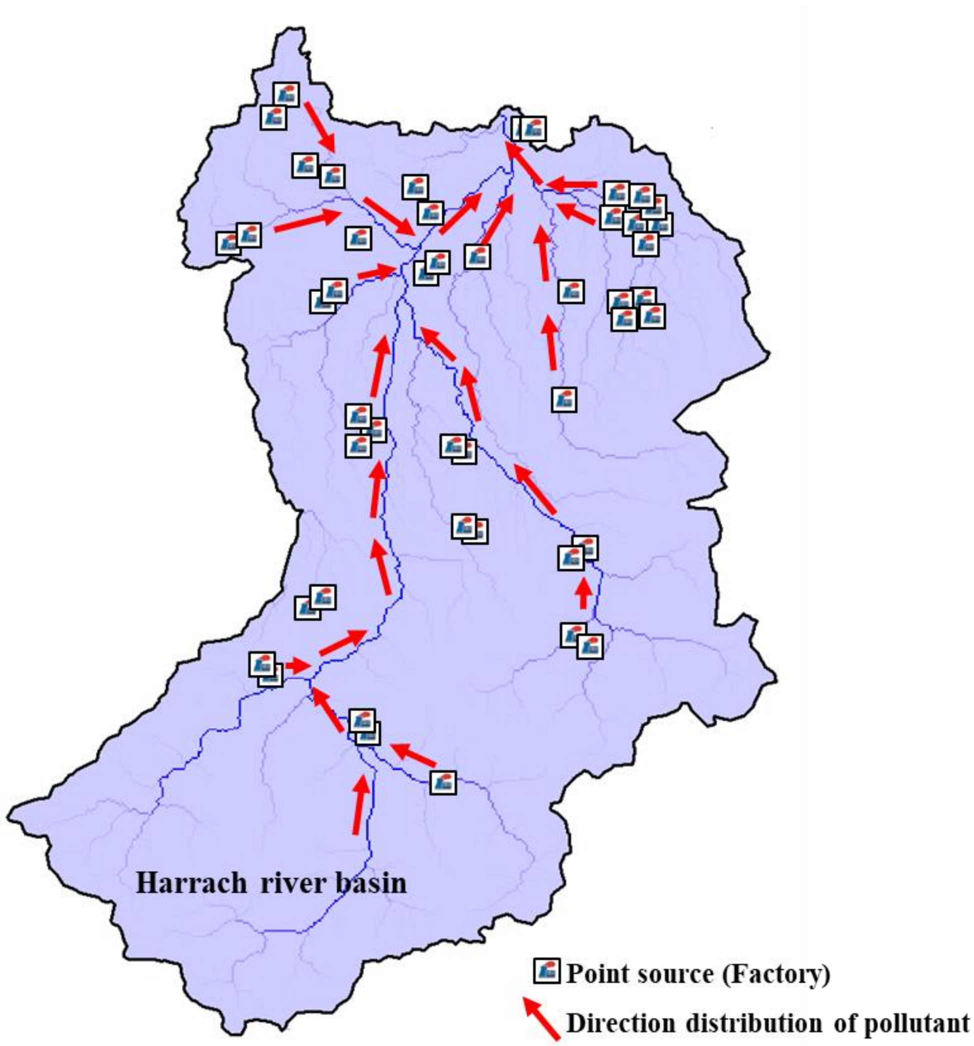


Fig. 4.5 Virtual presentation of the distribution of heavy metal concentration emissions along Harrach river basin

③ **Predict the results of different decisions in management strategies**

in this case, the model can calculate different results in scenarios in which we take different countermeasure to find the best solution of specific environmental problem. For example, we can assess the effect of point source load reduction (**Fig.4.6**). As in the method proposed in Chapter 2, we first assume that point source was removed from input data to compare the point source impacts to water quality. Second, we select the most important source as a target point (**Fig.4.7**), the simulation with the reduction of pollutant source load at the selected target source to certain amount by countermeasure such as wastewater treatment can predict the effect on water qualities at each point location. The procedure can provide a comprehensive assessment of the countermeasure effectiveness (**Fig. 4.8**).

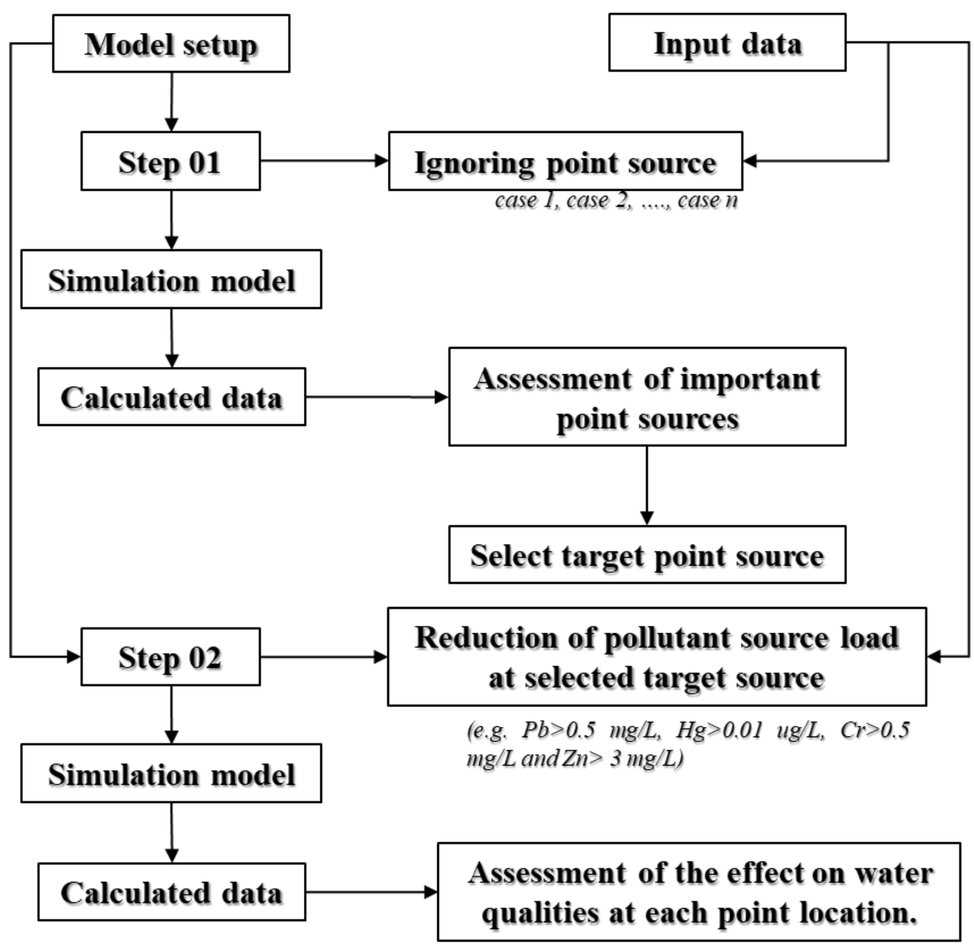


Fig.4.6 simple chart for assess the effect of point source load reduction.

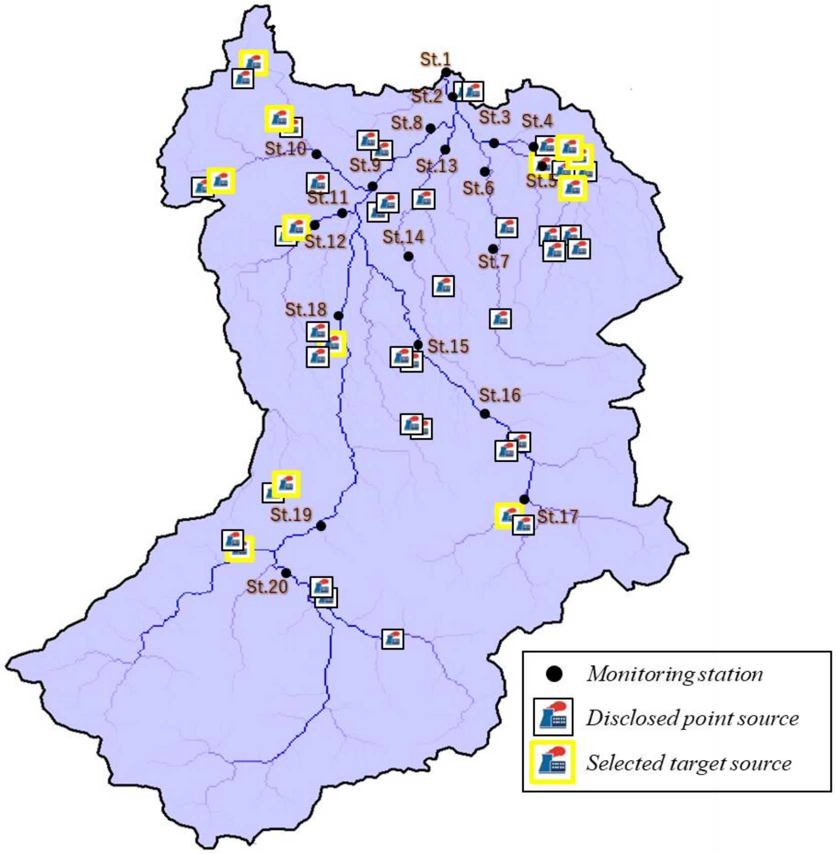


Fig.4.7 Virtual locations for selected target points source load.

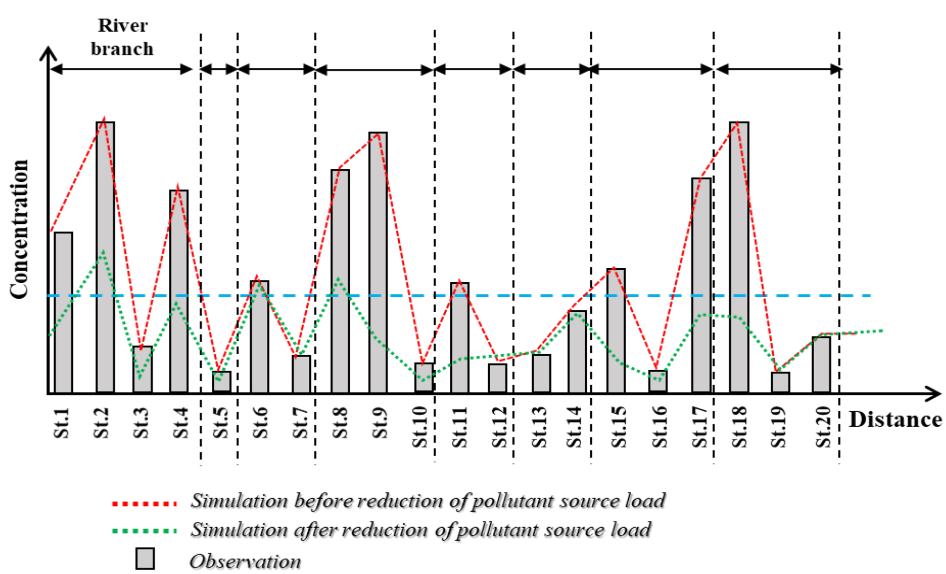


Fig.4.8 Virtual graphs for simulation results before and after reduction of pollutant source load.

Chapter 5

Conclusions

General conclusions

Recently, the interest in development of numerical models in water quality management has been increasing significantly. Many studies in the field of water quality modeling demonstrated the efficiency of hydrological models to predict the transport and fate of different pollutants in riverine system such as heavy metals. Therefore, the introducing of numerical models in water quality management policy would support to vulnerabilities in the monitoring structure.

The main objective of the thesis can be summarized as providing an evaluation methodology of heavy metals transport and pollution in riverine system for water quality management in developing countries, by adopting the numerical modeling as useful monitoring tool of heavy metal transport in aquatic systems.

The first chapter deals with literature reviews, in this chapter I have addressed three principal points: Heavy metals; Harrach River basin; Modeling; and water quality modeling and Heavy metal transport in river system.

The second chapter deals with the assessment of the Geo-CIRC model based on OOD for the simulation of multiple heavy metals concentration in stream water and sediment at different points along urban River. In this chapter, an evaluation of the Geo-CIRC model performance based on OOD for the simulation of contamination from Pb, Hg, Cr, and Zn contamination in Harrach River in Algeria, has been conducted. Furthermore, a methodology for the assessment of data quality control and the improvement of monitoring procedures was proposed by using the model to simulate different scenarios. The results showed the ability of Geo-CIRC model based OOD to predict the concentrations of multiple heavy metals with minimal input data. Also, the OOD increases the model's flexibility to allow the handling of many transportable materials. Therefore, multiple heavy metals can be numerically treated simultaneously. Likewise, the application of the Geo-CIRC model with systematic simulation scenarios in the present study can help to understand the riverine transport of multiple heavy metals which originate from industrial activities.

The third chapter deals with the proposition of simple approach for K_d model in order to improve the model accuracy for the simulation of heavy metal in sediment. It consists of two principal steps:

- **Step 01**: Generation of regression model of K_d of Pb considering physicochemical properties such as pH, SS, BOD and COD in riverine water. The obtained regression models in this study shows the influence of pH, SS and organic contents in determining the partitioning of Pb between sediment and stream water in Harrach River. The K_d values for Pb were reasonably estimated simply by introducing some water quality parameters i.e. pH, SS, COD and BOD.

- **Step 02**: Incorporating the presented K_d model with Geo-CIRC model to simulate Pb concentrations in sediment of Harrach River, Algeria. The results from the simulation incorporating K_d values changes were compared with those obtained from the simulation with constant K_d values. This comparison showed that incorporating of K_d model in simulation increases the model accuracy.

As I mention in chapter 4, the importance of this research highlights in terms of use of the Geo-CIRC model integrated with OOD to predict the transport of multiple heavy metal in riverine system. The application of the Geo-CIRC model in Harrach river case is the first work in its kind, as the modeling has never used to assess the contamination of rivers with heavy metal in Algeria before, whereas all the assessment studies were based on measured data. Therefore, the using of present simulation approach in monitoring of various heavy metals in river basins would be an important starting point for reliable monitoring system, in order to confront the problem of river pollution with heavy metal caused by industrial activities in Algeria.

Among the most prominent reasons for using Geo-CIRC model in monitoring system is providing better information for less cost and effort within a short time. The Geo-CIRC model can provide a comprehensive description of river contamination with using minimum amount of observation data, and supports monitoring efforts, while reducing the amount of necessary monitoring data and number of samples that must be analyzed for decision making. In addition, the model allows a predictive assessment of future situations after different strategies: for example, the model can use for assessing the impact of reducing the industrial effluent discharge. Or, Assessment of improved water quality after building of wastewater treatment plant.

Considering the outcomes in the chapters, this research study is having important progresses:

- The monitoring benefits from better technological support,
- Data processing tools are able to deal with less quantity of data,
- A simple approach with improved accuracy have been proposed.

Conclusions and recommendations:

[1] Geo-CIRC model based on OOD can be used in water quality monitoring of heavy metal transport in riverine system,

[2] Positive advantages of this model application are reducing costs, efforts and time.

[3] There is a possibility to conduct studies on other rivers and more accurately.

[4] Increasing of model accuracy when incorporating the K_d-model considering pH, SS and organic matter contents. However, further research is needed to test the possibility of modeling the transport of heavy metal using K_d model applications to include other elements than Pb.

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APPENDIX

Appendix I

Limit values of heavy metals concentrations in the industrial effluent to discharge in the urban network, according to the **Executive Decree No. 09-209 of June 11th, 2009**, laying down the procedures for granting authorization to discharge non-domestic wastewater into a public sewage network or a wastewater treatment plant. **"Maximum limit values for the content of harmful substances in non-domestic wastewater discharged into a public sewage system or a treatment plant."**

PARAMETERS	MAXIMUM LIMIT VALUES (mg/l)
Aluminum	5
Argent	0.1
Arsenic	0.1
Beryllium	0.05
Cadmium	0.1
Chrome trivalent	2
Chrome hexavalent	0.1
Chromates	2
Cooper	1
Cobalt	2
Cyanide	0.1
Tin	0.1
Iron	1
Magnesium	300
Mercury	0.01
Nickel	2
Lead	0.5
Zinc	2

Appendix II

Limit values of heavy metals concentrations in the industrial effluent to discharge in the natural aquatic system, according to the executive decree No. 06-141 of April 19th, 2006, **defining the limit values for discharges of industrial liquid effluents.**

PARAMETERS	MAXIMUM LIMIT VALUES (mg/l)
Aluminum	3
Argent	/
Arsenic	/
Beryllium	/
Cadmium	0.2
Total chromium	0.5
Chrome trivalent	/
Chrome hexavalent	/
Chromates	/
Total cooper	1
Cobalt	/
Cyanide	0.1
Total tin	2
Iron	3
Magnesium	/
Manganese	1
Total mercury	0.01
Total nickel	0.5
Total lead	0.5
Total zinc	3

Appendix III

Inventory of some industrial units existing in the study area and their severity of pollution with heavy metals.

Factory name	Activity	Treatment system of wastewater exist	Pollution level	Discharge network
ENPEC	Manufacture of starter accumulators	Exist of treatment system (decantation and pH neutralization)	Very polluted	Public network
ENAP	Manufacture of paint	• Exist of trois decantation basins • reuse of wastewater from cleaning in process	Very polluted	Public network
CATEL	Manufacture of cable	No information	Polluted	Public network
AVENTIS	Manufacture of medicine	No information	Not polluted	Public network
Hydrotraitment	Maintenance	No information	Very polluted	Public network
WINTHROP PHARMA SAIDAL	Manufacture of drugs	• Exist of three decantation tanks	Not polluted	Public network
CONCORDAL, SPA	Production of paint	• Exist of treatment system of liquid effluents	no information about the pollution level	Public network
GRANITEX NOUVEAUX PRODUITS	Production of concrete admixtures	• Decantation and reuse of process water	Not polluted	Public network
FAIENCERIE ALGERIENNE	Manufacture of floor slabs	• Exist of seven decantation tanks • reuse of 80% of water process	Not polluted	Public network
AGENOR SPA	Precious Materials company	• Exist of treatment system	Polluted	Public network
SARL, ACG	Galvanizing and metal fabrication	• Exist of treatment system (pH neutralization) • big part of wastewater is recycled	Polluted	Public network
EPE SACAR SPA	Paper processing	• Exist of decantation basins	Not polluted	Public network
ENMTP-UMB	Manufacture of public works equipment	• No information	Not polluted	Public network

EPIC ETUSA	Bus maintenance	• No system	Polluted	Public network
ETPH MOUNIB	Rental of construction machinery	• No system	Not polluted	Public network
TAIBA FOOD COMPANY	Beverage production	• Not system	Not polluted	Public network
SARL RAMY MILK COMPANY	Production of milk and dairy products	• No system	Not polluted	Public network
SARL RAMY BEVERAGE COMPANY	Beverage production	• No system	Not polluted	Public network
EPTP	Public works company	• Exist of treatment system of liquid effluents	no information about the pollution level	Public network
CENTRE LUBRIFIANT	Distribution and storage of lubricants	• Exist of system treatment of liquid effluents (decanter / oil separator)	no information about the pollution level	Public network
Abattoire El HARRACH	Slaughterhouse	• No system	Not polluted	Public network
ALFEL	Foundry	• No treatment exist for the wastewater from the cars cleaning	no information about the pollution level	Public network
ZET CERAM	Production of ceramic tiles	• Exist of two decantation basins	Not polluted	Public network
SPA GRANDS MOULINDAHM ANI LA BELLE	Cereals processing	• No treatment exist for the wastewater from the cars cleaning	Not polluted	Public network
NAFTAL, 116 A	Distribution and storage of hydrocarbons	• Exist of two hydrocarbons separator	no information about the pollution level	Public network
NAFTAL, 216 A	Distribution of oil products	• No treatment exist for the wastewater from the cleaning activities	no information about the pollution level	Public network
NAFTAL AVIATION	Marketing of aviation products	• Separation of water-hydrocarbon by physical decantation	Not polluted	Public network
ECFERAL	Production of industrial boiler	• Exist of treatment system	Not polluted	Public network
SEFLEX	Flexible packaging manufacturing	• Exist of treatment system	Not polluted	Public network

ENPC	Plastic transformation	• No information	Polluted	Public network
ENTREPRISE KEHRI DAHMANE	Treatment of hides and skins of cattle and sheep	• No information	Very polluted	Public network
ETABLISSEMEN T SEMMACHE AHMED	Skin treatment	• No information	Very polluted	Public network
RAFFINERIE D'ALGER	Refinery	• No information	Polluted	Public network
PRUI DELICE	Beverage manufactory	• No system exist	Not polluted	Public network
AUTO VERRE	Manufactory of all types of windshield	• Decantation pit	Not polluted	Public network
SARL ALGO FOOD	Coffee roasting production	• Exist of two decantation basins	Not polluted	Public network
GENERAL LAITERIES	Manufactory of flavored milk and beverages	• Exist of treatment system of liquid effluents (neutralization and decantation)	Not polluted	Public network
EPE ALPHYT	Formulation of phytosanitary product	• No system exist	Not polluted	Not connected with public network
SARL TECHNOFLEX	Cartridge transformation	• No system exist	Not polluted	Public network
SAVIAL MARBRERIE UNIT	Transformation and sale of marble	• Exist of settling tank	Not polluted	Not connected with Public network
SARL LAITIERE MYSTERE	Milk production	• Not system exist	Not polluted	Public network
SARL RAND FOOD FANY GLACES	Ice cream production	• No system exist	Not polluted	Public network
Ets KEHRI DAHMANE	Tannery	• No system treatment of liquid effluent exist	Polluted	Public network
EMB	Metal packaging manufacturing	• No information	polluted	Public network
BAG	Manufacture of gas cylinders	• No information	Polluted	Public network
SOACHLORE	Chlorine production	• No system of treatment exist	Very polluted	Public network

List of Publications

Journal Papers:

[1] Saadia BOURAGBA, Katsuaki KOMAI, and Keisuke NAKAYAMA: **“Assessment of distributed hydrological model performance for simulation of multi-heavy-metal transport in Harrach River, Algeria”**. Water Science & Technology, Vol. 80 (1), pp: 11–24. doi: 10.2166/wst.2019.250.

[2] Saadia BOURAGBA, Katsuaki KOMAI, and Keisuke NAKAYAMA: “**Empirical approach for modeling of partition coefficient on lead concentrations in riverine sediment**”. Journal of Environmental Science and Development (IJESD, ISSN:2010-0264) (accepted)

Reference papers:

[1] Saadia BOURAGBA, Katsuaki KOMAI, and Keisuke NAKAYAMA: “**Heavy Metals Transport Simulation by Physically Based Distributed Modeling Approach (Case Study: Harrach River, Algeria)**”. Journal of Japan Society of Civil Engineers, Ser. B1 (Hydraulic Engineering), Vol. 73, No. 4, I_1159-I_1164, 2017.

International Conferences:

[1] Saadia BOURAGBA, Katsuaki KOMAI, and Keisuke NAKAYAMA: **“Assessment of River Contamination with Various Heavy Metals Using Distributed Hydrological Model (Case Study: Harrach River, Algeria)”**. Water and Environment Technology Conference, JWET. July 2018. pp: 5. Johoku Campus of Ehime University, Matsuyama, Japan.

[2] Saadia BOURAGBA, Katsuaki KOMAI, and Keisuke NAKAYAMA: **“A preliminary study of the partitioning of heavy metals in water-solid phases for assessing Cr mobility within surface water”**. 3rd international symposium of river basin studies organized by River Basin Research Center, Gifu University, Gifu, Japan, 5th – 6th March 2019.

[3] Saadia BOURAGBA, Katsuaki KOMAI, Keisuke NAKAYAMA and Toshiyuki CHIBA: **"Mobility and partitioning of heavy metal in the basin of Mellah River, Algeria"**. Water and Environment Technology Conference, JWET. July, 2019. pp: 84. Suita Campus of Osaka University, Osaka, Japan.

[4] Saadia BOURAGBA, Katsuaki KOMAI, and Keisuke NAKAYAMA: **"Empirical approach for modeling of partition coefficient on lead concentrations in riverine sediment"**. 6th International Conference on Environment and Bio-Engineering (ICEBE 2020). Uji Campus, Kyoto University, Kyoto, Japan. January 19-22, 2020.

List of Achievements

[1] **"Best Presentation Award"** from the 3rd international symposium of river basin studies organized by River Basin Research Center, Gifu University, Gifu, Japan, 5th – 6th March 2019.

[2] **"Best Presentation Award"** from the 6th International Conference on Environment and Bio-Engineering (ICEBE 2020). Uji Campus, Kyoto University, Kyoto, Japan. January 19-22, 2020.

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