

Empirical Approach for Modeling of Partition Coefficient on Lead Concentrations in Riverine Sediment

Saadia Bouragba, Katsuaki Komai, and Keisuke Nakayama.

Abstract—Since a large part of heavy metals input in aquatic system accumulates in sediment, their concentrations in sediment are regarded as an important indicator of the heavy metal pollution of aquatic environment. The partition coefficient (K_d) is an empirical parameter that can represent the interaction of heavy metals at the sediment-water interface in aquatic system, however, it is not always stable with environmental conditions. Therefore, the introduction of K_d model with dominant physicochemical parameters would facilitate and improve the simulation of heavy metals concentrations in riverine sediment. The present study aims to develop a K_d model considering four physicochemical properties in stream water in order to simulate heavy metal concentrations in sediment of severely polluted urban rivers. Lead (Pb) concentrations in sediment of Harrach River, Algeria, were simulated using one-dimensional distributed hydrological model incorporating with presented K_d model. Multivariable equation of K_d model with physicochemical parameters (pH, suspended solid concentration (SS), chemical oxygen demand (COD) and biological oxygen demand (BOD)) was obtained from multiple regression analysis with observation data in various environmental condition. Hydrological simulations were tested with K_d model comparing to giving constant K_d . The numerical results agreed better with K_d model than with constant K_d , where the results accuracy increased from R^2 0.05 for constant K_d to R^2 0.67 for K_d model.

Index Terms—Distributed hydrological model, Harrach River pollution, heavy metal, partition coefficient modeling, regression analysis.

I. INTRODUCTION

Heavy metals are recognized as danger contaminants in aquatic environment, even at low concentrations [1]. These elements are natural components of rocks, however, the increase of their concentration level is originating from anthropogenic activity such as urbanization, industrialization [2]. The sediment is also related with heavy metal pollution, because a large part of the heavy metal input eventually accumulates in sediment [3]. Therefore, the monitoring of the accumulation of these pollutants in sediment is important for river water quality management.

The assessment based on numerical computational models by water resource planners, water quality managers, engineers and scientists is getting more and more popular in recent several decades [4]. Arguably, the numerical model is

a useful tool for predicting of heavy metal transport and fate in aquatic system. The partitioning of heavy metals between particulate and dissolved phases in polluted aquatic system is intricacy phenomenon that is strongly related to the environmental conditions. Unfortunately, this interaction makes the modeling of heavy metal in riverine system difficult and more complicated. However, it is possible to describe the interaction of heavy metal between solid and water by assuming that the concentration of the metal sorbed to the solid particle is proportional to the concentration of the metal in solution [5]. The K_d is an empirical parameter which depends on various factors, and it is commonly used for describing solid-solution interaction [6]. Some researchers have been able to estimate the K_d values of various heavy metals in term of different physicochemical properties using multiple regressions analysis. For example, Rene *et al.* 1997, [7] estimated the K_d values of various heavy metals by specifying the pH of the soil. Carlon *et al.* 2004, [8] generated multiple regression equations with the variables pH, to predict the K_d values of Pb. Therefore, it is possible to estimate the K_d values of heavy metals from various physicochemical characteristics and using it in the simulation of heavy metal concentration in sediment to improve the accuracy of the model.

In present study, we proposed an empirical multivariate regression model of K_d considering physicochemical properties (pH, suspended solid concentration (SS), and organic content) in riverine water to improve the estimation accuracy of K_d [9]. Where, the regression equations based on pH, SS, chemical oxygen demand (COD and biological oxygen demand (BOD) were derived to estimate K_d values of lead (Pb). Next, the concentrations of Pb in sediment of Harrach River, Algeria, which is severely polluted by many industries with heavy metals, were simulated by using the numerical model incorporating with the K_d model.

II. STUDY AREA DESCRIPTION

The study area is located in Harrach River Basin, one of the large rivers in Algeria, extending over 51 km from north to south and 31 km from east to west, an area of 1270 km² [10], [11]. Harrach River crosses the province of Algiers, the capital city of Algeria. This city is characterized by large industrial activities. Unfortunately, the industrial activities are almost being operated without any environmental controls, and causes the pollution of Harrach River with various pollutants [10].

According to Yoshida *et al.* 2005, [12], Harrach River is highly polluted with various heavy metals such as Cr, Pb, Hg, As, Cd and Zn, which are originated from the industrial activities.

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III. DATA PREPARATION

A. Watershed and River Network Generation

The digital elevation model (DEM) (obtained from CCGIAR-CSI consortium for special information) [13] data were used to calculate river network and watershed with ArcGIS. The river network and the watershed were simulated with a surface grid cell size of 100 m. The delineated river network comprises 5 branches numbered from 0 to 4 as shown in Fig. 1.

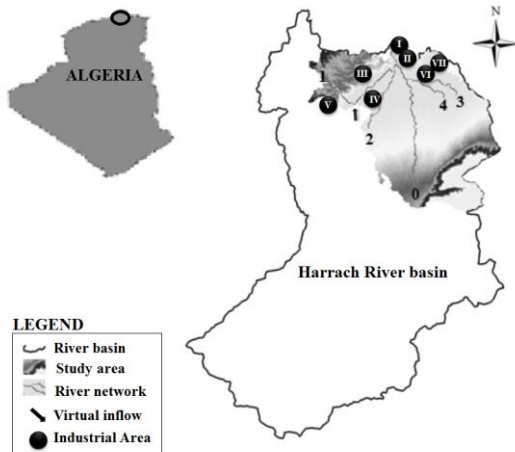


Fig. 1. Study area description.

B. Input Data

Pb was considered for calculation in Harrach River basin. Table I shows major input sources of Pb concentrations and flow rate (Q) data of wastewater discharged from factories at different points along the river network.

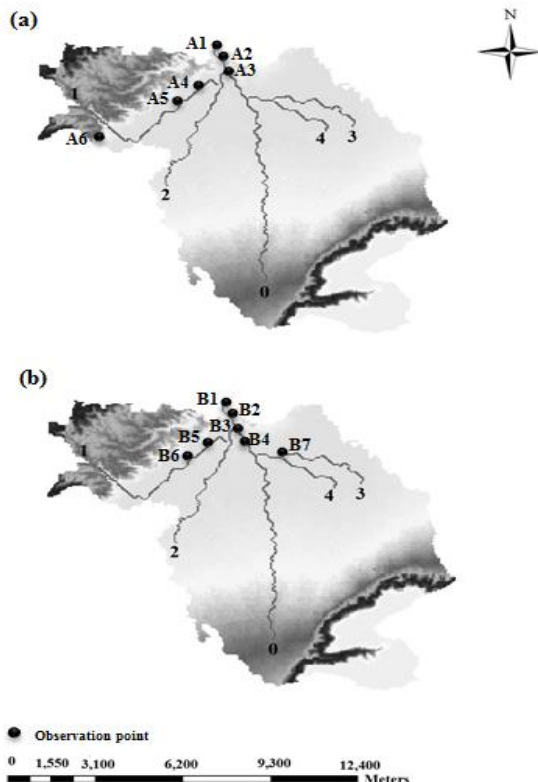


Fig. 2. Observation point's locations in the study area. (a) Point location for data shown in Table II, (b) Point location for data shown in Table III.

Fig. 2 show the locations of observation points presented in Tables II and III.

The data shown in Table II were used to calibrate and generate appropriate regression equation of K_d between $\log K_d$ of Pb and the physicochemical characteristics (pH, SS, COD and BOD), since are among indicators of pollution according to the general regulations for wastewater qualities in Algeria. Another dataset was applied for the verification of the K_d model (Table III). All the data using in current study are the outcomes of evaluation study carried out in 2004-2011 by the cooperation team between ONEDD (Observatoire National de l'Environnement et de Developpement Durable) and JICA (the International Cooperation Agency of Japan), on Harrach river pollution with various heavy metals caused by the industrial activities. The field sampling campaigns were conducted in 3/2005, 11/2006-7/2007, and 2/2010, along Harrach River at different points according to the program established by ONEDD and the experts of JICA.

TABLE I: INPUT DATA OF PB CONCENTRATIONS OF WASTEWATER DISCHARGED FROM FACTORIES [ONEDD 2005]

Factory name	Area location	Discharge location	Q (m ³ /day)	Pb (mg/L)
ENMTP	I	river 0	3	0.20
ENPC	II	river 0	/	0.27
Raff Alger	IV	river 1	7	0.51
EMB1	III	river 1	320	2.40
BAG	III	river 1	100	0.45
SOACHLORE	V	river 1	930	0
Est KEHRI	IV	river 1	50	0.23
Tan Semmache	VI	river 3	30	2.23
Tan KEHRI	VII	river 3	120	0.27
AGENORE	VI	river 3	3000	0.34
CATEL	VI	river 3	12	0.94
AVENTIS	VI	river 3	/	0.38
ENAP	VI	river 3	7	0
ENPEC	VI	river 3	150	37
Hydrotraitment	VI	river 3	/	22

TABLE II: PB CONCENTRATION IN WATER AND SEDIMENT, PARTITION COEFFICIENT AND THE PHYSICOCHEMICAL CHARACTERISTICS (PH, SS, BOD AND COD) [ONEDD 2006-2007]

	Pb in water (mg/L)	Pb in sediment (mg/kg)	K_d (L/kg)	pH	SS (mg/L)	BOD (mg/L)	COD (mg/L)
A1	0.6	137	228	7.48	18	110	15
A2	0.73	200	273	7.41	77	140	2825
A3	0.6	83	138	7.46	14	140	370
A4	0.54	86	159	8.04	77	56	130
A5	0.6	217	361	7.2	480	170	5400
A6	0.57	130	228	7.7	370	130	170

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

TABLE III: PB CONCENTRATION IN WATER AND SEDIMENT, PARTITION COEFFICIENT 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)	pH	SS (mg/L)	BOD (mg/L)	COD (mg/L)
B1	0.4	200	500	7.6	100	45	690
B2	0.6	83	138	7.5	77	140	370
B3	0.37	287	776	7.4	170	39	220
B4	0.89	170	191	7.9	1500	420	140
B5	0.53	144	2712	7.7	190	22	0.5
B6	0.57	130	228	7.9	510	130	330
B7	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.

IV. MODELS FORMULATION

The modeling procedures of flow and pollutant transport in riverine system is mainly based on solving the hydro-environmental equations [9]. The governing equations used are as following.

A. River Flow Model

The longwave equations have been applied to estimate water level and discharge [9]. The detail of the numerical model (GeoCIRC) to solve the water flow was indicated in previous papers [14]-[17].

B. Heavy Metal Transport Model

The distribution of dissolved heavy metals in rivers can be modeled by the advection equation [18]. The one-dimensional advection equation was used to simulate the dissolved heavy metal concentration as follows:

$$\frac{\partial c}{\partial t} + \frac{\partial Q}{\partial X} = (A_a + B_a) \quad (1)$$

where C = dissolved heavy metal concentration, Q = discharge, 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 relation [18]:

$$A_a = \frac{q_L C_a}{\Delta x} \quad (2)$$

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

C. Partition Coefficient

The interactions of heavy metals between solid-liquid phases have been simplified by assuming that the concentration of the metal sorbed to the solid particle is proportional to the concentration of the metal in solution. The ratio defined as K_d [5] is expressed as,

$$K_d = \frac{C_{sed}}{C} \quad (3)$$

where C_{sed} = heavy metal concentration in sediment (mg/kg), C = heavy metal concentration in water (mg/L), K_d = partition coefficient (l/kg).

The concentration of heavy metal in sediment can be derived from (4) as follows:

$$C_{sed} = K_d \times C \quad (4)$$

D. Model Setup

The hydrodynamic module of the Geo-CIRC model developed by [19] was set up to simulate the flow field and heavy metal concentrations in Harrach River.

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 m³/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. 1). Since at some factories near rivers 0 and 3, the discharge data were not available (Table I), however these points are located in a highly industrialized area where there are many factories. Therefore, point source loads from these areas were assumed to be 10^4 m³/day and 10^3 m³/day at rivers 0 and 3, respectively, in order to represent the river discharge at the downstream end of river 0 [9].

The transport of Pb concentration in water was modeled with assuming that the interaction of metal concentrations between stream water and sediment in equilibrium state [9]. Then, the concentrations of Pb in sediment were calculated using equation (4). Two cases of simulation with K_d values for Pb were conducted. In case 1, we used the average K_d values estimated by [9] ($K_{dPb}=400$ (L/kg)) in the same river basin. In case 2, we used the variable K_d values at each point location; those were estimated from the empirical model in terms of pH, SS, COD and BOD as mentioned below.

For the empirical approach, the regression equation should have included physicochemical parameters that can explain most of the variation in K_d and that are easily obtainable for model's usability. K_d values were calculated from equation (4) using the observed data shown in Table II. Multiple regression equation of measured $\log K_d$ values to four physicochemical parameters (pH, SS, COD and BOD) shown in Table II were obtained as K_d model calibration by means of the Mathematica.

$$\log K_d(Pb) = 7.0013 - 0.00359641 \text{BOD} + 0.000178826 \text{COD} - 0.572996 \text{pH} + 0.000504306 \text{SS} \quad (5)$$

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

We have used the coefficient of determination (R^2) for evaluation of the goodness of fit. Also we validated empirical equations to the other observation data shown in Table III.

Likewise, we have generated alternative regression models with ignoring one parameter for each scenario in order to assess the importance of each selected parameter for the prediction of K_d values.

$$\log K_d(Pb) = 2.22092 - 0.000181906 \text{BOD} + 0.0000413731 \text{COD} + 0.000217953 \text{SS} \quad (6)$$

$$\log K_d(Pb) = 2.91552 - 0.00126562 \text{BOD} + 0.0000495889 \text{COD} - 0.0835693 \text{pH} \quad (7)$$

$$\log K_d(Pb) = 3.11673 + 0.0000307791 \text{COD} - 0.114511 \text{pH} + 0.000257957 \text{SS} \quad (8)$$

$$\log K_d(Pb) = 8.27116 - 0.00430179 \text{BOD} + -0.728905 \text{pH} + 0.00062743 \text{SS} \quad (9)$$

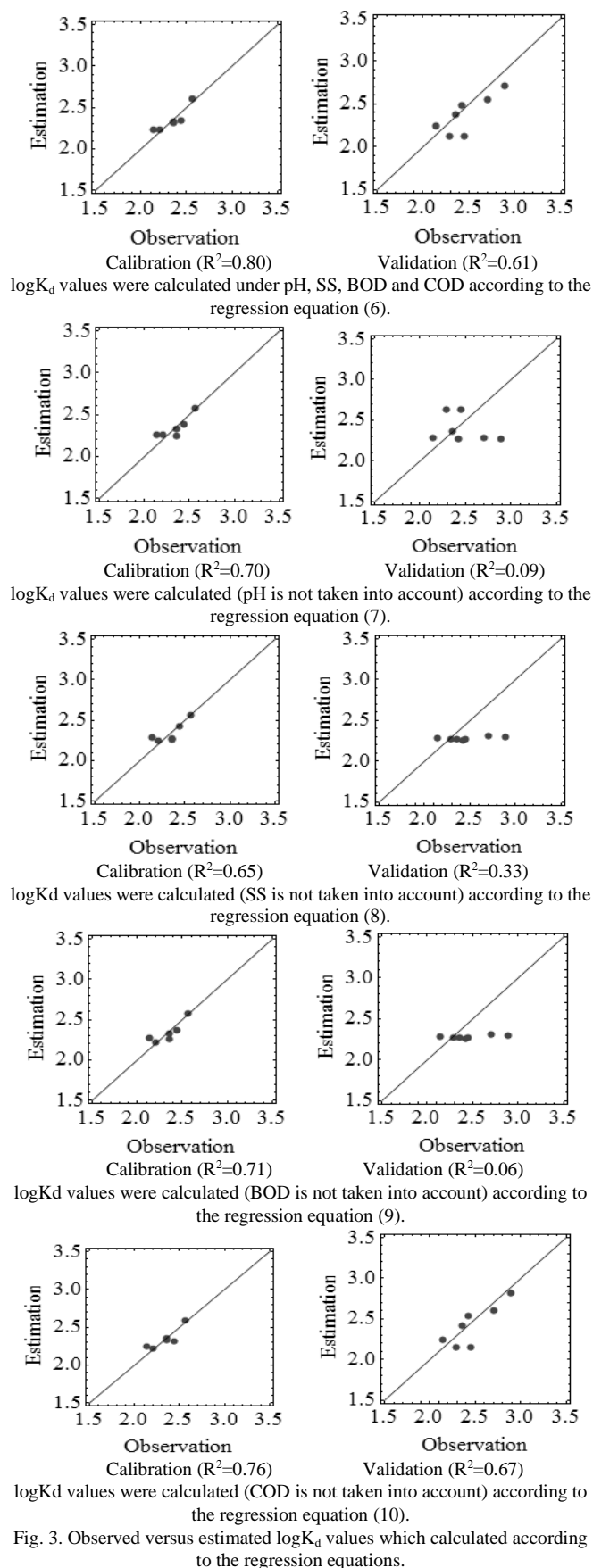
V. RESULTS AND DISCUSSION

A. Multiple Linear Regression

Fig. 3 show the calibration and validation of K_d model in case of equations (5) to (9).

In calibration, the left panels of Fig. 3, K_d values agreed well in equation (5) with observation ($R^2=0.80$). In contrast, R^2 values were slightly worse in case of equations (5) to (9),

i.e., 0.70, 0.65, 0.72, and 0.77, when pH, SS, BOD and COD is not taken into account in each case, respectively.



In validation, the right panels of Figs. 3, we have applied the regression equations to the data shown in Table III. The estimation of K_d values was relatively successful ($R^2=0.61$)

when taking into account all the selected physicochemical properties in equation (5). Moreover, it is found that COD was not significant parameter because it is slightly improved rather than affected when diminishing COD ($R^2=0.67$).

COD is the total concentration of all chemicals (organic and inorganic) in the water that can be oxidized, whereas, BOD is the total concentration of OM that bacteria can oxidize [20]. The low importance of COD (Figs. 3) can be attributed relatively to the weak of inorganic content-dependency compared to organic content.

According to [21], the high pH-dependent adsorption results from the chemistry of organic contents surface. Also, about 2% to 3% of the total solids in SS represents a particulate organic coating, providing to the surface important characteristics in the exchange of trace metals between solid-water phases. Furthermore, in aquatic system SS have pH-dependent surface characteristics, which affecting its reaction with certain functional organic contents groups [21].

The results published by [7], showed that the pH is the most influenced factors in determining K_d values of various heavy metals, whereas dissolved organic carbon also had an important role in the variation of K_d values. Carlon *et al.* 2004, [8] view that the application of chemometric methods to include the OM effects on metal partitioning in model seems promising, although their regression results showed that is not expected a significant model improvement by including of additional variables with pH. Therefore, using both of pH, SS, as well BOD and COD (as indicators of OM) might be significantly contributed to the variation of K_d models.

B. Simulation Results of Heavy Metal Concentration in Sediment

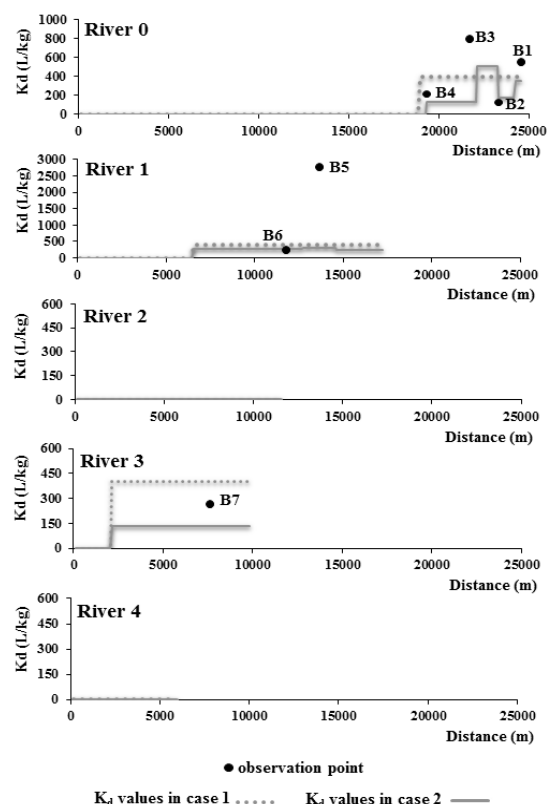


Fig. 4. K_d distribution in the downstream direction of each river branch.

Fig. 4 shows the distributions of K_d in the downstream direction of each river branch in cases 1 and 2. Here, K_d in case 2 were estimated from observation data in Table III by using equation (5) as mentioned above. K_d in rivers 0 and 3 were different in cases 1 and 2 while K_d in the other rivers were almost the same magnitude.

Fig. 5 shows the distributions of Pb concentrations in sediment in the downstream direction in cases 1 and 2. Here, Pb concentrations in sediment were numerically simulated by using constant K_d in case 1 and K_d model in case 2, respectively.

Fig. 6 shows the correlations of Pb concentration between observation data shown in Table III and simulation in cases 1 and 2. The results agreed better in case 2 with observed than in case 1. This means that the model accuracy successfully increased when we incorporated the K_d model using equation (6), i.e., $R^2=0.05$ for cases 1 and $R^2=0.67$ for case 2, respectively (Fig. 6).

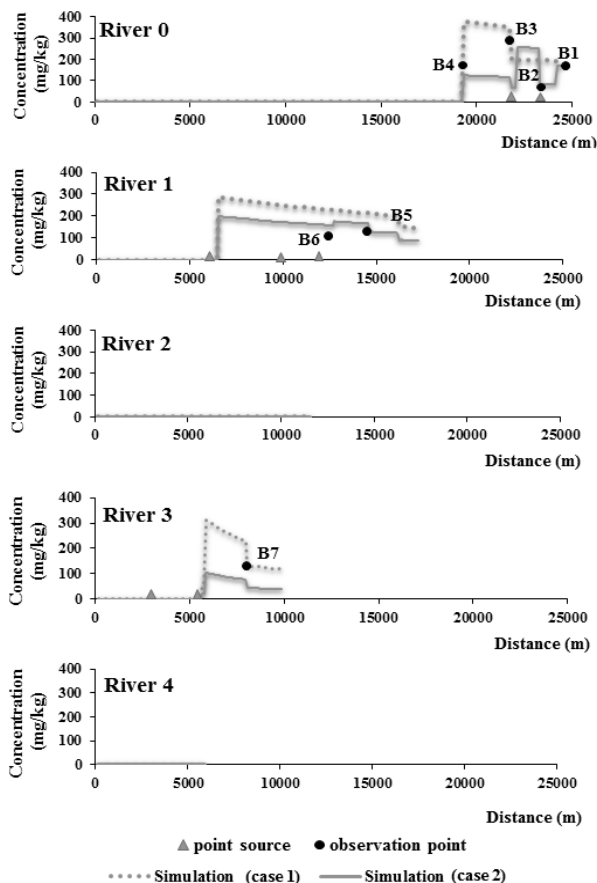


Fig. 5 Pb concentrations pattern in sediment in the downstream direction of each river branch.

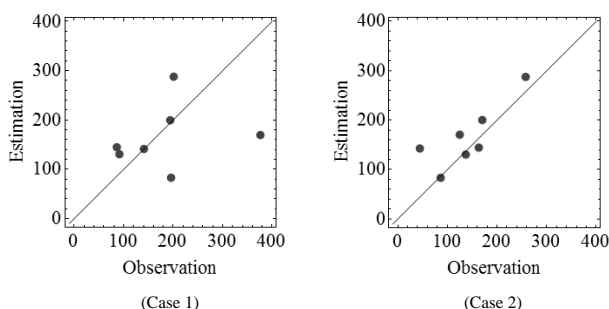


Fig. 6. Observed versus simulated concentration (mg/kg) of Pb in sediment.

Bouragba *et al.* 2019, [9] provided a methodology predicting heavy metal concentrations of various in sediment using constant value of K_d that calculated from the measured data, however this assumption of K_d values relatively affected the results accuracy, because the K_d is not always constant, it is affecting by elements properties as well the characteristics of solid and water phases [7]. Falconer & Lin 2003, [22] used salinity to model the partitioning coefficient of heavy metals in the Mersey estuary and they found that simulation results were in good agreement with observed data. The importance of K_d model in present study is therefore to introduce a procedure that related to pH, SS and OM contents values to K_d for estimation of the varied K_d values as accurately possible. Thereupon, for heavy metal modeling, K_d model would be relatively helpful for accurate simulation of heavy metals concentrations in sediments.

Consequently, it seems possible to improve the simulation accuracy of Pb concentrations in sediments by using K_d model calculated from BOD, COD, pH, and SS. However, there are limitations in using this regression model for hydrological model in present 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.

VI. CONCLUSION

In this paper, details were given for distributed hydrological modeling of heavy metal concentrations in the sediment using partition coefficient. The model was successfully applied to estimate Pb concentrations in sediment of Harrach River in Algeria. The regression models were useful in estimation of the potential variables of the partition coefficient with physicochemical properties changes, therefore introducing these changes in simulation increased the result accuracy of the model.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

All authors contributed the study design and they contributed towards the data preparation as well as the model application; Saadia Bouragba and Katsuaki Komai contributed significantly towards drafting the manuscript, verifying the findings of the work and the related interpretation of the results; all authors had approved the final version.

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