



## Modelling of arsenic retention in constructed wetlands



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### HIGHLIGHTS

- As retention processes in constructed wetlands were implemented in RCB-ARSENIC.
- As precipitation and sorption, and root oxygen release were included.
- Data from two planted prototypes were used to test the model.
- The simulated data closely matched the data measured in all evaluated cases.
- The RCB-ARSENIC model has provided reasonably good response values.

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### ABSTRACT

A new model was developed in order to simulate the most significant arsenic retention processes that take place in constructed wetlands (CWs) treating high arsenic waters. The present contribution presents the implementation phases related to plants (arsenic uptake and accumulation, root arsenic adsorption, and root oxygen release), showing the first simulation results of the complete model. Different approaches with diverse influent configurations were simulated. In terms of total arsenic concentrations in effluent, the simulated data closely matched the data measured in all evaluated cases. The iron and arsenic species relationships, and the arsenic retention percentages obtained from simulations, were in agreement with the experimental data and literature. The arsenic retention efficiency increased whenever a new phase was implemented, reaching a maximum efficiency range of 85–95%. According to the quality of the obtained results, it can be considered that the implementation of all steps of RCB-ARSENIC provided reasonably good response values.

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### 1. Introduction

The presence of high arsenic (As) concentrations in drinking water has been declared a major risk to human health in many parts of the world (Lièvreumont et al., 2009). The best available candidate technologies for the treatment of drinking water are principally based on physical and chemical processes: filtration, coagulation, adsorption, ion exchange and reverse osmosis. They have proved highly efficient in reducing As concentrations in water. However, their most important drawback is that their operation and maintenance costs are sometimes prohibitive. The need therefore exists to develop low cost technologies to tackle the problem.

An economically viable and feasible alternative treatment option is the constructed wetland (CW) technology (Lièvreumont

et al., 2009). For over four decades CWs have been used for the treatment of a wide set of waters (Vymazal, 2011), such as landfill leachate (Bulc, 2006) and mine drainage waters (Sobolewski, 1996). Their application in As removal from water is relatively recent. Rahman (2009) showed that CW systems were highly efficient in removing As from a synthetic wastewater. Moreover, Alarcón-Herrera et al. (2012) showed that these systems could also be highly efficient in removing As from a synthetic water for drinking purposes. Much has been elucidated in the last few decades regarding As speciation and the important parameters and processes that affect the speciation and mobility of As under different conditions (Lièvreumont et al., 2009; Smedley and Kinniburgh, 2002). However, there is still a great lack of knowledge concerning the performance of CW systems in the treatment of high As waters (Lizama et al., 2011), mainly due to the fact that As retention in these systems involves a large number of physical and biochemical processes that take place simultaneously (Olmos-Márquez et al., 2012; Singhakant et al., 2009).

Modelling, in particular the use of mechanistic mathematical models, has become a powerful tool to gain a greater understanding

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of the performance of water treatment systems (Rivas et al., 2008). The development of mechanistic models describing processes in CWs is relatively recent. Only a few models are able to simulate the biochemical transformation and degradation processes that occur in horizontal subsurface flow CWs (HSSF CWs) for the treatment of urban wastewater (Langergraber, 2008; Llorens et al., 2011a,b; Ojeda et al., 2008; Rousseau, 2005; Samsó and García, 2013). However, no mechanistic models have yet been developed that are capable of simulating the As retention processes that occur in HSSF CWs in the treatment of high As waters.

The aim of the present study was to adapt RetrasoCodeBright (RCB) (Saaltink et al., 2003), in order to simulate the most significant As retention processes that take place in HSSF CWs treating groundwater. The first and second implementation phases (which consider the reactions describing aqueous complexation, As precipitation, and As sorption processes in the granular media) of the resulting RCB-ARSENIC 2D simulation model were completed and provided reasonably good response values (Llorens et al., 2013). The main objective of this contribution is to present the processes implemented in the next phases (third, fourth, and fifth) in RCB (that means those reactions describing As uptake and accumulation in plants, as well as those describing As adsorption to plant roots and root oxygen release from plants), and to show some first simulation results of the complete model.

## 2. Methods

### 2.1. The RCB code

The RCB code is a powerful modelling tool that has been successfully applied in various hydrogeological studies. The code formulates the flow problem through a multiphase approach that includes porous media composed of solid grains, water and gas. It enables the simulation of the reactive transport of inorganic dissolved and gaseous species in non-isothermal saturated and unsaturated problems by finite elements (Saaltink et al., 2003). The transport of solutes in the water column is modelled by means of advection, dispersion and diffusion, together with chemical reactions. Advective flux and dispersive/diffusive fluxes are computed by means of Darcy's and Fick's laws, respectively. Gas-liquid interactions are assumed to be sufficiently fast with respect to flow to reach equilibrium (represented by Henry's law).

For the numerical solution of the reactive transport equations, RCB uses the direct substitution approach (Saaltink et al., 2004), which consists of substituting the chemical reactions in the transport equations and solving them simultaneously, usually by means of the Newton-Raphson method. The code can use one-, two- or three-dimensional finite element grids.

Llorens et al. (2011a,b) and Mburu et al. (2013) demonstrated that RCB could be used for HSSF CWs simulation satisfactorily. Accordingly, RCB was modified to include the most significant reactions involved in the mobility of As in HSSF CWs.

### 2.2. Model adaptation

The resulting 2D simulation model, called RCB-ARSENIC, was obtained by means of incorporating new reactions related to As retention in RCB (flow and reactive transport model based on mass balance equations). The reactions considered for the model adaptation were those set out by Rahman (2009): precipitation, adsorption, uptake and accumulation in plants. The root oxygen release from plants was also considered. In this respect, the implementation of the processes was structured in five phases. The first phase involved the implementation of the precipitation and aqueous complexation reactions in RCB; while the second, third, fourth

and fifth phases were related to the sorption processes in the granular media, the As uptake and accumulation processes into the plants, the sorption processes in the plant roots, and the root oxygen release from plants, respectively.

#### 2.2.1. Implementation of precipitation processes

Six mineral precipitation-dissolution reactions were included in the model: scorodite, mansfieldite, quartz, k-feldspar, muscovite, and montmorillonite-mg. The precipitation-dissolution reactions of the two first minerals follow a mineral-solution equilibrium, while the reactions of the remaining minerals follow a kinetic law (Llorens et al., 2013). The decision was taken to consider only the minerals related to the type of HSSF CWs considered to be simulated (groundwater treatment and granular media composition), which is described in Section 2.3.

The As-related aqueous complexation reactions considered in the RCB-ARSENIC model are almost instantaneous and can therefore be effectively considered equilibrium reactions (Llorens et al., 2013). Reactions related to iron (Fe) were also included in the model, since the groundwater, taken as an example for treatment, had some concentrations of this metal (Section 2.3).

For the sake of simplicity, the physical transfer of oxygen from the atmosphere to the water was included. More details can be found in Llorens et al. (2013).

#### 2.2.2. Implementation of sorption processes in the granular media

The implementation of As sorption processes in the granular media consisted of adding the sorption rates to the reaction term of the RCB mass balances (Eq. (1)):

$$\frac{\partial C}{\partial t} = -q\nabla C + \nabla(D\nabla C) + r \quad (1)$$

where  $C$  is the component concentration [ $M M(H_2O)^{-1}$ ],  $t$  is the time [T],  $q$  is the Darcy's flux [ $L T^{-1}$ ],  $D$  is the dispersion-diffusion coefficient [ $L^2 T^{-1}$ ], and  $r$  is the reaction term [ $M T^{-1} L^{-2}$ ]. Accordingly, the first term refers to the advective flux [ $M T^{-1} L^{-2}$ ], the second term refers to the diffusive-dispersive fluxes [ $M T^{-1} L^{-2}$ ] and the third term refers to the reaction term [ $M T^{-1} L^{-2}$ ].

Due to the RCB architecture, the sorption processes were defined following a kinetic rate law formulation based on Monod kinetics in accordance with Eq. (2):

$$r = \sigma_m \prod_{m=1}^{N_m} f_m \quad (2)$$

where  $r$  is the rate,  $\sigma_m$  is the reactive surface [ $m^2/m^3$  rock],  $N_m$  is the number of terms of the expression, and  $f_m$  is a term of the expression. Each  $f_m$  can be one of the three types described in Eqs. (3)–(5), depending on their function in kinetic rates (p-order, catalysis and inhibition factors, respectively).

$$f_m = km \prod_{i=1}^{N_i} C_i^{p_{im}} \quad (3)$$

$$f_m = \frac{\prod_{i=1}^{N_i} C_i^{p_{im}}}{km + \prod_{i=1}^{N_i} C_i^{p_{im}}} \quad (4)$$

$$f_m = \frac{km}{km + \prod_{i=1}^{N_i} C_i^{p_{im}}} \quad (5)$$

where  $km$  is the rate constant,  $N_i$  is the number of species in solution,  $C_i$  is the concentration of the  $i$ th species solution and  $p_{im}$  is a parameter.

The As sorption processes in the granular media were described in one unique reaction as Eq. (6) shows, in which the equilibrium constant  $\log$  is  $-30.0$ .

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