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Understanding the retention and fate prediction of copper ions in single and competitive system in two soils: An experimental and numerical investigation

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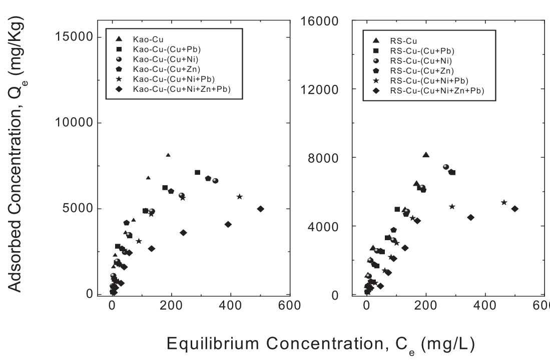
HIGHLIGHTS

- Single and competitive interaction of copper in two soils
- Contaminant fate prediction of copper-soil interaction varies with competition.
- Retardation factors different for Freundlich (R_F) isotherm and Langmuir (R_{La})
- Fate prediction theorms do not consider multiple contaminant interaction

GRAPHICAL ABSTRACT



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ABSTRACT

The concept of sponge city has become very popular with major thrust on design of waste containment systems such as biofilter and green roofs. Factors that may influence pollutant ions retention in these systems will be soil type and also their interactions. The study investigated single and competitive interaction of copper in two soils and its influence on the fate prediction. Freundlich and Langmuir nonlinear isotherms were selected to quantify the retention results. Series of numerical simulations were conducted to model 1 D advection-dispersion transport for the two soils and analyse the role of isotherms. The results indicated that contaminant fate prediction of copper-soil interaction based on the two non-linear isotherms was different for both single and that in competition. Retardation factor obtained from Freundlich (R_F) isotherm predicts more than Langmuir (R_{La}). This observation is more explicit at the higher range of equilibrium concentration. Fate prediction based on retardation value obtained from retention isotherms exhibited some anomalous trends contradicting the experimental findings due to inherent assumptions in governing equations. The necessity to have an approximate assessment of contaminant concentration in the field to effectively use contaminant retention results for accurate fate prediction is highlighted here. The study is important for modellers in design or analysis of biofilter system (sponge city), where multiple ions tend to exist in waste water.

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1. Introduction

Subsurface and ground water contamination by heavy metal copper originating from natural soil sources is a serious global issue with detrimental impacts on human wellbeing and ecological health (Clothier et al., 2010; Gupta and Agate, 1988; Chandra, 1980; Chotpantara et al., 2011). Finding appropriate remediation plan for the prediction of the behaviour of copper in the soil and incorporating the environmental consequences in the ecosystem is of utmost necessity. The fate of copper is controlled by retention/release reactions, precipitation and complexation. The importance of geochemical processes on the migration of contaminants through the soils is well documented (Rowe, 1989; Rowe and Booker, 1990; Soller and Berg, 1992). Transport and retention are the two important mechanisms that govern the presence of a contaminant in the geosphere with respect to time and space. The soil-contaminant interaction influences the partition of the metal in the liquid and soil phases and are responsible for its mobility and bioavailability in the system. However, one of the challenges for making accurate fate prediction is the estimation of the flux of contaminants through soil to the groundwater under varying geologic, hydrologic, and chemical conditions (Cantrell et al., 2003). For complex geo chemical process subjected to varying boundary conditions in space and time, numerical modelling is a crucial tool. For such complex problems, experimental studies over sufficiently long distances and/or time periods is not always feasible (Vanderborght et al., 1996; Zhang and Selim, 2007). For accurate prediction of the fate of contaminants in the geoenvironment, the contaminant retention characteristics obtained from the laboratory were often used as input parameters in the mathematical models (Parker and Genuchten, 1984; Selim, 1992; Shukla et al., 2002). However, it can be noted that for most of the studies the determination of input retention parameters is primarily based on a single model contaminant. Extensive studies were conducted on the interaction of single metal ions with various minerals and soils (Vasudevan et al., 2002; Vega et al., 2006; Scheinost et al., 2001; Trivedi et al., 2001; Gomes et al., 2001;). In the past, studies were done to evaluate the use of sustainable materials such as natural materials (Bordoloi et al., 2017; Bordoloi et al., 2018) or vegetation (Das et al., 2017; Gadi et al., 2017; Garg et al., 2017a) for bio-remediation of soil. Apart from this, attempts have also been made to use litter manure and biochar for sorption of contaminants. Uchimiya et al. (2010) employed sorbents of a broiler litter manure under different degree of carbonization for sorption of copper ions. In their study, they concluded that pH and surface area affects retention capacity of biochar. In another study, Uchimiya et al. (2011) observed that different soil types (presence of negatively charged soil particles) could also affect the sorption isotherms of copper ions. Chen et al., 2011 evaluated potential of biochars obtained from pyrolysis of wood and corn straw for retention of copper ions in competition with zinc ions. They concluded that retention of copper ions is hardly affected by presence of zinc ions.

The interaction of single ion with soils may not adequately portray the retention behaviours of heavy metal in the presence of multiple ions. The contamination in soil is influenced widely by an array of competitive species rather than a single ion. The various interactions in these multicomponent or multiple-ion systems significantly impact the fate and transport of the pollutants, and competition for sorption sites on soil matrix surfaces (Zhang and Selim, 2012; Brusseau et al., 2012).Thus, considering competitive sorption is an important part of predicting contaminant transport. Such studies are critical for designing efficient waste containment facilities for municipal and industrial wastes (Rowe and Booker, 1985; Yong et al., 2001; Zhang et al., 2005; Poly and Sreedeep, 2011). The retention of Cu^{2+} may get influenced by the presence of other heavy metals (Covelo et al., 2004; Gomes et al., 2001; Jalali and Moharrami, 2007; Serrano et al., 2005; Spark and Wells, 1995). Pb^{+2}, Ni^{+2} and Zn^{+2} are some of the most commonly encountered heavy metals found at hazardous-waste sites (Sims and Sommers, 1985; Wuana and Okieimen, 2011). Such influence need to be systematically investigated. The objective of this study is to investigate the fate of heavy metal copper based on the retention characteristics obtained from single and when in competition with other heavy metals Pb, Ni and Zn. The various multiple contaminant systems are divided into binary: Cu + Ni, Cu + Zn, Cu + Pb); ternary (Cu + Ni + Pb) and quarternary (Cu + Ni + Zn + Pb). The study was conducted for two different types of soils. The results of single and multiple contaminant retention are quantified and compared using sorption isotherms (Elkhatib et al., 1991; Hooda and Alloway, 1998; Villegas et al., 2004; Serrano et al., 2005; Buragohain and Sreedeep, 2013). The breakthrough curves (BTCs) to the convection-dispersion equation was conducted by CXTFIT curve-fitting software.

2. Theoretical background

2.1. Sorption isotherms

Contaminant retention or sorption characterization for a particular soil-contaminant interaction is carried out based on laboratory tests and with the help of mathematical models known as isotherms (Foo and Hameed, 2010, Sharma and Reddy, 2004; Lair et al., 2007). The retention isotherm is defined as the relationship between the contaminant retained on the soil (Q_e in mg/kg) and the soil pore water concentration (C_e in mg/L) at equilibrium (Wang and Qin, 2005; Aluyor and Oboh, 2009). Batch equilibrium tests are generally employed to estimate the sorption properties of the soil which is mathematically represented by isotherms that are typically linear at low contaminant concentrations and nonlinear at higher contaminant concentrations. It can be noted that linear isotherm was generally used owing to its simplicity but the distribution coefficient (K_d) for estimating the retardation factor used in modelling contaminant transport may only be realistic at low contamination levels. At high level of contamination, the simplistic approach could lead to contaminant transport being significantly underestimated (Coles, 2002). An inherent drawback of this approach is its empirical nature. This is because of the variation in a K_d value cannot be confidently estimated beyond the certain range of chemical conditions under which it was measured. The utility of any K_d measurement is good only for that specific set of conditions. Hence, for more accurate estimation of retardation and contaminant transport, nonlinear isotherms are best suitable. Freundlich and the Langmuir models have been used in various studies of Cu^{+2} retention (Petruzzelli et al., 1985; Temminghoff et al., 1994; Yuan and Lavkulich, 1997). They are represented by Eqs. 1 and 2 respectively. Freundlich isotherms assume infinite sorption sites; hence, the maximum limit is not achieved. However, this is not true in practice, because there will be only finite number of sorption sites. The Langmuir isotherm is a more realistic nonlinear isotherm that accounts for a limited number of sorption sites and takes into account the decrease in partition coefficient when the adsorption sites are partially saturated with contaminants:

$$Q_e = \frac{Q_m K_L C_e}{1 + K_L C_e} \quad (1)$$

$$Q_e = K_F {C_e}^N \quad (2)$$

Where C_e is the equilibrium concentration (mg/L), Q_e is the adsorbed concentration (mg/kg), K_L is the retention constant related to the binding energy (L/mg), and Q_m is the maximum contaminant adsorbed by solid (mg/kg), K_F is the Freundlich partition coefficient, and N is constant.

2.2. Retardation factor for different isotherms

The retardation factor (R) is one of the coefficients that describe and measure the migration potential of particular components in groundwater. This coefficient indicates the soils ability to restrain the transport