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Review

The remediation of heavy metals contaminated sediment

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ABSTRACT

Heavy metal contamination has become a worldwide problem through disturbing the normal functions of rivers and lakes. Sediment, as the largest storage and resources of heavy metal, plays a rather important role in metal transformations. This paper provides a review on the geochemical forms, affecting factors and remediation technologies of heavy metal in sediment. The in situ remediation of sediment aims at increasing the stabilization of some metals such as the mobile and the exchangeable fractions; whereas, the ex situ remediation mainly aims at removing those potentially mobile metals, such as the Mn-oxides and the organic matter (OM) fraction. The pH and OM can directly change metals distribution in sediment; however oxidation–reduction potential (ORP), mainly through changing the pH values, indirectly alters metals distribution. Mainly ascribed to their simple operation mode, low costs and fast remediation effects, in situ remediation technologies, especially being fit for slight pollution sediment, are applied widely. However, for avoiding metal secondary pollution from sediment release, ex situ remediation should be the hot point in future research.

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1. Introduction

Nowadays, heavy metals originating from anthropogenic activities are frequently detected in sediments and water columns of river/lake, which cause a considerable number of the world's rivers/lakes severely contaminated [1–4]. Heavy metals pollution had gradually become a major concern worldwide.

In aquatic environment, heavy metal is usually distributed as follows: water-soluble species, colloids, suspended forms and sedimentary phases. However, unlike organic pollutants, natural processes of decomposition do not remove heavy metals. On the contrary, they usually are enriched in sediment by organisms or some other compounds. In some conditions, more than 99% of heavy metal entering into river can be stored in river sediments in various forms [5]. However, heavy metals cannot fix in sediment forever. With the variation of the physical–chemical characteristics of water conditions, part of these fixed metals will re-enter the overlying water and become available to living organisms. Thus, sediment often acts as both carriers and potential sources for metals in aquatic environment [1]. Heavy metals usually possess significant toxicity to aquatic organisms, and then affect human health through food chain. Therefore, investigating the transformation and distributions mechanisms of heavy metal in sediment becomes necessary.

The main purpose of this paper is to provide a review on the geochemical forms, affecting factors and remediation technologies of heavy metal in sediment, depending on literatures and practical working experiences. Only based on these, the optimization of remediation technologies fitting for polluted sediment can be achieved.

2. Metals species in sediment

In sediments, metals can be bound to various compartments in different ways: occluded in amorphous materials; adsorbed on clay surfaces or iron/manganese oxyhydroxides; presenting in lattice of secondary minerals like carbonates, sulfates or oxides; complexed with organic matter (OM) or lattice of primary minerals such as silicates [6,7]. Since each form has different remobilization potential, and then affects its respective bioavailability and toxicity, the measurement of total metal may not be able to provide exact information about the characteristics of pollution [8].

To clearly investigate the toxicity of heavy metal to aquatic biota, in the past decades, different sequential extraction procedures for partitioning the metals bound to various mineral components had been developed [9]. Based on these, the community Bureau of Reference of the Commission of the European Union initiated a major effort to harmonize these extraction procedures. Their works produced the definition of an extraction protocol (the BCR protocol) and a purely operational definition of sequential fractionation. For enlarging the research scopes, depending on the different partition conditions among various metals, some assistant sequential extraction procedure were also established and popularized. Some primary characteristics of them are shown in Table 1.

In all of these sequential extraction methods, the four steps method mainly established by Tessier et al. won the most popular application [6]. This method partitions the heavy metals into five fractions: extractable and exchangeable, carbonate bound, iron and manganese oxides bound, organic matter bound and residual metal.

Exchangeable fraction, usually extracted with magnesium chloride solution or sodium acetate solution (1 M) at pH 8.2 for 1 h, refers to the metals directly adsorbing on sediments [16]. Through some typical sorption–adsorptions processes, these metals can be exchangeable and are in equilibrium with the ionic content in water. Generally, this fraction is usually used to represent the environmentally available components.

Carbonates fraction, continuously extracted with NaOAc or HOAc solution (1 M) at pH 5.0 for 5 h, mainly refers to the metals that are precipitated or co-precipitated with carbonate. This fraction is sensitive to pH variations.

Fe–Mn oxides fraction includes the soluble metal oxides/hydroxides under slightly acidic pH as well as the metal associated with reducible amorphous Fe–Mn oxyhydroxides, which was extracted with 25% (v/v) acetic acid containing some $NH_2OH{\cdot}HCl$ at 96 °C for 6 h. This fraction can be dissolved with oxidation–reduction potential (ORP) varying.

Organic fraction may be associated with various forms of organic material such as living organisms, detritus or coatings on mineral particles through complexation or bioaccumulation process. It is extracted mainly with 0.02 M nitric acid and 30% hydrogen peroxide at pH 2.0 and 85 °C. This kind of metals can exist in sediment for longer periods, and can also be released with OM decomposition.

Residual fraction, namely the metals still remained in sediment after the above extraction procedures, usually presents as consolidated oxides, co-precipitates, and strongly held complexes, which keeps relatively stable and does not show significant transformation in various conditions.

Normally, the summation of the mobile and the exchangeable fractions can be used to assess the environmentally available components. The fractions bound to Mn oxides and organic material are supposed to represent the potentially mobile component under changing conditions, which are reviewed as the most important components in sediments for metals binding. While the residual fraction represents the more stable metal forms associated with anthropogenic or geogenic components, the influence of which to ecological system is much less than the others in major conditions.

3. Influence factors for heavy metal distributions

It has been validated that each environmental factor presents unique influence on metals distribution in sediment. The influences of some factors, such as pH, ORP and OM are more crucial than the others. Only a slight variation of them, the distributions of metals would be producing some significant variations. Correspondingly, some other factors (e.g. salinity, temperature) can only alter metals distribution to a less extent. In this paper, only those relatively important factors are introduced.

3.1. *The influence of pH values*

The pH is a key parameter controlling heavy metal transfer behavior in sediment. Normally, with pH decreasing in sediment, the competition between H^+ and the dissolved metals for ligands (e.g. OH^-, $CO_3{}^{2-}$, $SO_4{}^{2-}$, Cl^-, S^{2-} and phosphates) becomes more and more significant. It subsequently decreases the adsorption abilities and bioavailability of the metals, and then increases the mobility of heavy metal. Sometimes, only with a few lower of pH units, the fixation percentage of heavy metals on sediment particles may range from almost a 100% to none [17]. In sediment, due to the OM degradation and the acid volatile sulfide oxidation, the pH of sediment usually decreases from the neutral in the initial to acid, sometimes even decreasing to pH 1.2, which often results in some metals being released into water again even under stable water conditions [15,18].