Occurrence Characteristics and Ecological Risk Assessment of Heavy Metals in Sewage Sludge

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Abstract. Sewage sludge, as the main product of sewage treatment processes, contains relatively high concentrations of potentially toxic heavy metals. If it is not thoroughly treated or controlled, it will cause serious impact on the environment and human health. The aim of this review is to briefly generalize the occurrence characteristics of heavy metals in sewage sludge and give an overview of sludge standards.

1. Introduction
With the rapid development, China's water consumption is also increasing and a large amount of wastewater is discharged into the environment every day. Moreover, China has stringent requirements and standards for the effluent quality of wastewater treatment plants. As of the end of September 2015, a total of 3,830 wastewater treatment plants have been built in cities and towns across the country, and the sewage treatment capacity has reached 162 million m³/d. The sewage sludge has exceeded 30 million t/year (water content 80%). According to the statistics of the Ministry of Housing and Urban-Rural Development, as of the end of 2014, about 56% of the sludge was disposed of by building materials, incineration, fertilizer, sanitary landfill, etc. Contaminants in the sludge can enter the soil through various means, thereby jeopardizing human health through the food chain. There have been many studies on heavy metals in sewage sludge. The purpose of this review is to briefly introduce the sources and hazards of heavy metals, the occurrence level of heavy metals in sludge, the standards for sludge treatment and disposal and the contamination assessment methods of heavy metals in sludge.

2. Sources of heavy metals in sewage sludge and their hazards
The heavy metals in domestic sewage are mainly from the use of some metal-containing chemicals, such as cosmetics, detergents, etc[1, 2]. Heavy metals in industrial wastewater are the main sources of heavy metals in sludge. The content of heavy metals in sewage sludge dominated by industrial wastewater is significantly higher than that of municipal sewage. In addition, there are various types of heavy metals produced in different industries. For example, leather wastewater contains a large amount of Cr, and wastewater from metallurgy and electroplating plants contains a large amount of Cu, Ni, Cd. The mining, electronics, chemical and mechanical industries are also prone to discharge heavy metal-containing wastewater, while food, plastics and pharmaceutical industries emit less or no heavy metals.

High concentrations of heavy metals can destroy the normal physiological processes of plants, leading to a decline in yield and quality. They can also become a threat to human health after entering the food chain[3]. Lead pollution in soils and sediments has led to elevated levels of lead in children's blood worldwide[4], causing adverse physiological and neurological effects on children. Due to the
properties of toxicity, persistence, and bioaccumulation, some trace heavy metals (Cd, Cr, As, Hg, Pb, Cu, Zn, Ni) have been listed as priority pollutants by the US Environmental Protection Agency[5].

3. Occurrence level of heavy metals in sewage sludge

Many scholars have carried out researches on the heavy metal of sewage sludge from various sewage treatment plants in many different regions. In addition, researches on heavy metals in sewage sludge in various provinces and cities of China have also been carried out [6-11]. More than 50%-80% of the heavy metals in the sewage are concentrated in the produced sludge through various ways such as bacterial absorption, surface adsorption and co-precipitation[10]. The studies found that in urban wastewater treatment plants, elements such as Zn and Cu are generally high in sludge, and heavy metals such as Cd, Hg and As of high toxicity are relatively low [12, 13]. The total content of most heavy metals is significantly different. The magnitude of the change is relatively large [14]. In addition, the heavy metals in the sludge also showed significant differences with time and season. Due to factors such as the different influent water sources and wastewater treatment processes, the content of the same metal in different wastewater treatment plants is different[13]. While analyzing the total content of heavy metals, many researchers also analyzed the content of heavy metals in different forms. The most commonly used method in the literature is the BCR three-step continuous fractionation extraction method [15]. This method divides the sludge into four forms, which are acid dissolved/exchangeable, reducible, oxidizable and residual fractions.

4. Overview of sludge treatment and disposal standards

The content of heavy metals in sludge is one of the criteria for use or disposal. The standards for heavy metals in sewage sludge have already existed in many countries[16]. In 1984, China issued the first national sludge standard for the control of pollutants in agricultural use (GB 4284-1984). Now the current sludge standards are classified into mandatory standards and recommended standards based on the implementation nature of the standards. The US sludge standards mainly refer to standards for the use or disposal of sludge promulgated by the EPA (40 CFR Part 503). The standard was published in February 1993 and mainly includes three subparts: land application, surface disposal and incineration. The European Union's sludge agricultural use (Directive86/278/EEC) is the main directive for the EU on sludge. Some Member States have implemented stricter ceiling values for heavy metals based Directive86/278/EEC. In addition, the relevant standards for sludge treatment and disposal in the European Union include the Directive75/442/EEC on hazardous waste, the Directive2000/76/EC on the incineration of waste, and the Directive 99/31/EC on the landfill of waste. In 1972, Germany passed the first Waste Disposal Act (AbfG). Subsequently, the sludge method (AbfKlaeV) was developed, which regulates the properties of sludge used for agricultural, forestry and horticultural land. At the same time, the sludge standard is also the specific implementation of the EU Agricultural Soil Protection Directive (Directive86/278/EEC). There are three main sludge standards in the UK: The sludge (use in agriculture) regulations, the waste control regulations and the waste collection and disposal regulations[17]. The limit values for heavy-metal concentrations of different countries in sludge for agriculture use are shown in the table 1.

Table 1. Limit values for heavy-metal concentrations in sludge for use in agriculture (mg/kg of dry matter)

| Standard (Country) | As | Cd | Cu | Pb | Hg | Ni | Cr | Zn |
|-------------------|----|----|----|----|----|----|----|----|
| GB 4284-1984 (China) | pH<6.5 | 75  | 5  | 250 | 300 | 5  | 100 | 600 | 500 |
| GB 4284-2018 (China) | pH>6.5 | 75  | 20 | 500 | 1000 | 15 | 200 | 1000 | 1000 |
| GB 4284-2018 (China) | Grade A | 30  | 3  | 500 | 300  | 3  | 100 | 500  | 1200 |
| GB 4284-2018 (China) | Grade B | 75  | 15 | 1500| 1000 | 15 | 200 | 1000 | 3000 |
| 40 CFR Part 503 (US) | 75  | 85 | 4300| 840 | 57  | 420 | -   | 7500 |
| Directive86/278/EEC (EU) | 20-40 | 1000-1750 | 750-1200 | 16-25 | 300-400 | -   | 2500-4000 | |
| (Sweden) [15] | 2   | 600 | 100 | 2.5 | 50  | 100 | 800 |
| AbfKlaeV (Germany) | 10  | 800 | 900 | 8   | 200 | 900 | 2500 |
5. Ecological risk assessment of heavy metals in sewage sludge

5.1 Geo-accumulation Index (Igeo)
Geo-accumulation index (Igeo) was proposed by German scholar Muller in 1969. It has been widely used as a quantitative indicator for studying the degree of heavy metal contamination in sediments or sludge. It can effectively evaluate the pollution level of pollutants in the environment. It is defined as:

\[ I_{geo} = \log_2 \left( \frac{C_n}{1.5B_n} \right) \]

Where \( C_n \) is the examined concentration of metal \( n \) in sewage sludge, \( B_n \) is the geochemical background value of metal \( n \), and 1.5 is a constant. \( C_n \) and \( B_n \) are in the same concentration unit.

5.2 Potential Ecological Risk Index (RI)
The potential ecological risk index method is used by Swedish scientist Hakanson to evaluate the heavy metal pollution in sediments. The formula is as follows:

\[ E_r^l = T_r^l \frac{C_n}{B_n} \]

\[ RI = \sum_{n=1}^{n} E_r^l \]

Where RI is the ecological risk assessment value of heavy metals, \( E_r^l \) is the potential ecological risk value for a certain metal. \( T_r^l \) represents the toxic response coefficient for the certain metal, \( C_n \) is the examined concentration of the certain metal, and \( B_n \) is the geochemical background reference value. \( C_n \) and \( B_n \) are in the same concentration unit.

6. Summary
This review focuses on the occurrence characteristics and environmental risk assessment of heavy metals in sewage sludge. The heavy metals in sewage sludge mainly come from domestic and industrial wastewater, especially some metal-containing industries. The heavy metals do not degrade with time, causing potential risk on human health and environment. Some studies have already reported the concentration levels in different regions. The concentrations of heavy metals differ due to the different sources of influent water and wastewater treatment processes. For the purpose of safe disposal and land application, limit values for heavy-metal concentrations in sludge have been established in many countries. In order to further understand the risk of heavy metals in sludge, contamination assessment methods have been used, including geo-accumulation index and potential ecological risk index.

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