Components of sludge from municipal wastewater treatment plants and its evaluation for land application in JiUJiang city of China

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Abstract. Municipal sewage sludge has a potential for widespread application on land, but the implementation of new standards for the agricultural use of sludge in China support a cautious approach. The organic matter, nutrients, and heavy metals in sewage sludge from four municipal wastewater treatment plants in Jiujiang City of China were continuously monitored. The feasibility for its land use was analyzed from three aspects such as sludge nutrients, heavy metal pollution levels, and potential ecological risks in soil. The average content of Cd, Cu, Zn, Cr, Ni, Pb, Hg and As in the sludge of Jiujiang City are 1.85, 305.25, 764.48, 170.57, 42.45, 42.27, 0.59, 28.79 mg·kg-1, which are less than or close to mean value of heavy metals in China (except As), and all meet the minimum standards of heavy metal content in the sludge according to Chinese Quality of sludge from municipal wastewater treatment plant (GB24188-2009). The maximum content of Cu, Zn, Cr, and As exceed the control standards of pollutants in sludge for agricultural use, and the maximum value of Cu and Cr are close to the limit of landscaping standards. The average contents of organic matter, total nitrogen, total phosphorus, total potassium, and total nutrients in sludge are 385.16, 34.14, 11.05, 7.87, 53.06 g·kg-1, respectively. The organic matter content meets the limit value (no less than 200 g·kg-1) of Chinese Control standards of pollutants in sludge for agricultural use (GB 4284-2018), and the total nutrients content meets certain content limit (no less than 40 g·kg-1) of total nutrients in China's organic fertilizer. The single-factor pollution index and Nemerow's synthetic pollution index methods were used to assess the pollution of heavy metals. The heavy metal pollution indexes (PI) are in descending order: Cd > Cu > Zn > Cr > As > Ni > Pb > Hg. The PI shows that Cd and Cu are in strong pollution level, Zn is in moderate pollution level, and As, Ni, Pb, Hg are in absolute safe level. The potential ecological risk levels of heavy metal are ranked in the following order: Cd> Cu> As> Ni> Zn> Cr > Pb. The monomial potential ecological risk coefficients reveal that Cd is in extremely strong ecological risk, Cu is in medium ecological risk and other heavy metals are in slight ecological risk. The results indicate that Cd and Cu are the main contributors to the pollution and potential ecological risks of heavy metals in soil, to which more attention should be paid during sludge disposal. Being rich in organic matter and inorganic plant nutrients， sewage sludge in Jiujiang City may be widely used on soil, but availability of potential toxic metals exceeding the limit value often restricts its uses. Before being harmlessly treated, the sludge in Jiujiang City should not be used on agricultural land and may be applied with highly caution on landscaping and land improvement, but it can be used as cover materials for co-landfilling.

Keywords: China's Municipal wastewater treatment plant (WWTP); Sludge components; Land application; Heavy metals; Potential ecological risk.

1. Introduction

Treatment and disposal of sewage sludge from municipal wastewater treatment plants have become a bottleneck that restricts the healthy development of the entire wastewater treatment industry. At the end of 2021, 4592 wastewater treatment plants have been established in cities throughout China, with a wastewater treatment capacity of reaching 247 million m3/day. The production of dry sludge in China have reached to 1.62 million tons/year, converted into wet sludge (moisture content 80%) up to 81.1 million tons/year. The sewage sludge from municipal wastewater treatment plants is rich in nutrients such as organic matter, nitrogen, phosphorus and potassium, as well as trace elements necessary for plant growth including Calcium, Iron, Sulfur, Magnesium, Zinc, Copper, Manganese, Boron, and Molybdenum. Land application of sewage sludge can increase the organic matter content of soil and improve the soil's chemical, physical and biological properties [1,2]. Developed countries such as the European Union and the United States have developed and established a relatively complete and scientific technical standard system [3,4], where the application rate of sewage sludge in agriculture is rather high. For example, the application rate was 45% on average for the 27 EU countries (2010), 36% for the United States (2011), 55% for Australia (2012), 12% for Japan (2014), 10% for New Zealand (2009), 79% for United Kingdom (2012), 30% for Germany (2015), 8% for Greece (2012) and 25% for Sweden (2014) [5]. Contrastively, in China the land application of sewage sludge has been restricted by the relevant regulations and standards, as well as factors like the properties and outlets of the sludge [6]. There are three types of pollutants that cause the environmental issues in land use, namely the pathogens, heavy metals and organic matter in sewage sludge. Among them, heavy metals have always been one of the major factors restricting land application. Although sewage sludge is rich in organic and inorganic plant nutrients and can thus partially replace fertilizers, its application is often constrained by the potentially toxic metals [2,7]. He et al. [8] found that some heavy metals in the sewage sludge exceeded the standards seriously. They concluded that the sewage sludge was unsuitable for land application, or even unsuitable as the landfill covering soil. In contrast, based on the soil environmental capacity estimates of heavy metal elements, Ma et al. [9] speculated that the heavy metal problem with sewage sludge would not become a limiting factor for its land application. This paper attempts to monitor the sludge components of four wastewater treatment plants in Jiujiang City for one consecutive year. Based on the test results, a feasibility analysis is carried out concerning the land application of sludge components, with a view to providing a reference for the harmless treatment of municipal sewage sludge and the rational utilization of lands.

2. Materials and Methods

2.1 Sources of sewage sludge

The sewage sludge sample was dewatered sludge from four representative wastewater treatment plants (WWTPs) in Jiujiang City(hereinafter referred to as A, B, C, and D). WWTP A adopted the CAST technology and had a daily wastewater treatment capacity of 300,000 t, which received domestic sewage mainly from the new urban and suburban areas. WWTP B adopted the oxidation ditch technology and had a daily wastewater treatment capacity of 100,000 t, which was mainly responsible for treating domestic sewage from the old urban districts. With a daily wastewater treatment capacity of 50,000 t by CASS technology, WWTP C received the domestic sewage and partial light industrial wastewater from the urban-rural junction areas. As for WWTP D, it adopted the SBR technology and had a daily wastewater treatment capacity of 3,000 t, which was mainly responsible for treating campus domestic sewage.

2.2 Sludge sampling and pretreatment

Sludge samples were taken once per month and 2-3 kg each time during 2021-2022. The collected sludge samples were air-dried under the natural conditions of well ventilation and low light, ground with agate mortar, passed through a 100-mesh nylon sieve, and then stored in sealed bags for later use.

2.3 Analytical testing methods

Nutrient components in the sewage sludge were tested in accordance with Chinese Determination Methods for Municipal Sludge in Wastewater Treatment Plants (CJ/T 221-2005).

Specifically, the organic matter, total nitrogen, total phosphorus, and total potassium were determined by the gravimetric method, the alkaline potassium persulfate digestion-UV spectrophotometry, the sodium hydroxide melting-molybdenum antimony anti-spectrophotometry, and the inductively coupled plasma atomic emission spectrometry following microwave high-pressure digestion, respectively.

Meanwhile, heavy metals in the sewage sludge were tested in accordance with Chinese Determination Methods for Municipal Sludge in Wastewater Treatment Plants (CJ/T 221-2005). Specifically, Zn, Cu, Pb, Ni, Cr, Cd, and As were determined by the inductively coupled plasma atomic emission spectrometry following microwave high-pressure digestion, whereas Hg was determined by the atomic fluorescence spectrometry following atmospheric pressure digestion.

2.4 Evaluation methods

2.4.1 Heavy metal pollution evaluation for sludge land application

a. Single-factor pollution index method: As a commonly used method worldwide, the single-factor index method evaluates the pollution degree of a certain heavy metal pollutant in sewage sludge based on the comparison and classification between measured and standard values. Relevant computational formula is as follows:

$$P_i = \frac{C_i}{S_i} \quad (1)$$

Where P_i denotes the environmental quality index of a heavy metal i; C_i denotes the measured content of the heavy metal i (mg·kg-1); and S_i denotes its environmental quality standard value (mg·kg-1). In this study, the risk screening values (pH≤5.5) for agricultural land pollution as stipulated in Chinese Soil Quality Control Criteria for Soil Pollution in Agricultural Lands (Trial) (GB 15618-2018) were adopted, which were Cd (0.3), Hg (1.3), As (40), Pb (70), Cr (150), Zn (200), Ni (60), and Cu (50), specifically.

b. Nemerow synthetic pollution index method: The Nemerow synthetic pollution index method can reflect the pollution status of various heavy metal pollutants in soil comprehensively, and is capable of highlighting the pollution levels of heavy metals that cause serious pollution. Relevant computational formula is as follows:

$$P_N = \sqrt{\frac{(P_{i,\max})^2 + (P_{ave})^2}{2}} \quad (2)$$

Where P_N denotes the Nemerow synthetic pollution index; $P_{i,\max}$ denotes the maximum value of single pollution index; and P_{ave} is the average value of single pollution indices for all pollutants. According to the Nemerow synthetic pollution index method, the heavy metal pollution in soil can be classified into five levels.

Potential ecological risk evaluation for heavy metals in sludge

Potential ecological risk index method, as one of the most widely used methods for soil and sediment quality evaluation around the world, takes comprehensive consideration into the toxicity of heavy metal elements, the sensitivity to heavy metal pollution, and the difference in regional background value between heavy metal elements. It also gives a quantitative classification of potential ecological risk levels for heavy metal elements. Relevant evaluation formula is as follows:

$$RI = \sum_{r}^{m} E_r^i = \sum_{i}^{m} T_r^i . C_f^i = \sum_{i}^{m} T_r^i . \frac{C^i}{C_n^i} \quad (3)$$

Where RI denotes the potential ecological risk index of multiple heavy metals in soil; C^i denotes the measured concentration of heavy metal pollutants (mg·kg-1); and C_n^i is the background value of soil environment in the selected Jiangxi regions [10], as shown in Table 1. Besides, C_f^i denotes the ecological risk index of individual heavy metal; T_r^i denotes the toxicity response parameter of pollutant; and E_r^i is the potential ecological risk of individual heavy metal. In Table 1, the toxicity response coefficients of various heavy metal pollutants are presented [11].

Table 1 Reference value and toxicity coefficient of heavy metals in soil (mg·kg^{-1})

Heavy metal	Cd	Cu	Zn	Cr	Ni	Pb	As
C_n^i	0.10	20.8	69.00	48.00	19.00	32.10	10.40
T_r^i	30	5	1	2	5	5	10

3. Results and Discussion

3.1 Main nutrients in sewage sludge

Sewage sludge in municipal WWTPs contains rich organic matter and plant nutrients. In this study, the organic matter and nutrients in sewage sludge from four typical WWTPs in Jiujiang City were tested for 12 consecutive months, and the test results were calculated for arithmetic average and standard deviation (see Table 2). Respecting organic matter, total nitrogen and total phosphorus, the sludge from three WWTPs A, B, and C did not differ greatly. The concentration ranges of organic matter, total nitrogen and total phosphorus were 260.66-400.18 g·kg-1, 12.91-36.79 g·kg-1, and 6.96-11.90 g·kg-1, respectively. In comparison, for WWTP D, the concentration ranges of organic matter, total nitrogen and total phosphorus were 394.35-633.28 g·kg-1, 57.13-74.79 g·kg-1, and 14.98-19.69 g·kg-1, exhibiting much higher values than the remaining three WWTPs. This was primary because the sample of WWTP D was the thickening tank sludge, while the samples of the other three plants were the dewatered sludge. Another reason was that the WWTP D was a campus treatment plant, which featured short wastewater transport distance, and low content of inorganic matter like silt in the sewage sludge. Respecting total potassium, the sludge from WWTPs A and B exhibited higher values, while those from WWTPs C and D exhibited lower contents.

Table 2 Average content of organic matter and nutrients in sewage sludge (g·kg^{-1})

WWTP	Statistics	Organic matter	Total nitrogen	Total phosphorus	Total potassium
A	Average value *	332.20±33.11	19.67±3.28	10.31±0.62	12.77±0.81
	Concentration range	284.04-385.76	12.91-28.21	8.51-11.90	9.28-14.75
B	Average value *	305.62±22.99	30.20±1.94	9.17±0.75	10.29±0.71
	Concentration range	260.66-364.58	25.47-36.79	7.11-10.91	9.18-12.96
C	Average value *	317.23±48.81	22.09±1.83	7.97±0.55	4.17±0.34
	Concentration range	266.75-400.18	18.15-28.65	6.96-9.69	3.44-5.06
D	Average value *	585.59±46.70	64.61±3.16	16.76±0.95	4.25±0.72
	Concentration range	394.35-633.28	57.13-74.79	14.98-19.69	2.68-6.43
Average concentration		385.16	34.14	11.05	7.87

* Standard deviation: data size n = 12; confidence level P = 95%, t = 2.201.

Based on substantial sludge samples, the scholars and institutions of China had tested and statistically analyzed the organic matter and nutrients in sewage sludge nationwide separately in 2003, 2008, and 2009 (see Table 3). In respect of organic matter, the content ranged between 280-411.5 g·kg-1 in the sludge in China. For the sludge from four studied WWTPs in Jiangxi of China, the average content was 385.16 g·kg-1, which was basically within the above range, and was rather close to the values of Shanxi of China [12] and Guangxi of China [13] as well. Respecting nutrient components, the total nitrogen, total phosphorus and total potassium contents in sludge for Jiangxi were 34.14, 11.05, and 7.87 g·kg-1, respectively, which were rather close to the average values of China in 2003 and 2008. Nevertheless, marked differences in total phosphorus content were found from China's 2009 value (22.6 g·kg-1), Guangxi (36.26 g·kg-1) and Shaanxi (3.11-7.48 g·kg-1), which might be associated largely with the regional lifestyle differences. According to the comparison across countries, the organic matter content of sludge was the highest for the United States, reaching 534 g·kg-1, while was the lowest for India, which was 55.20-126.00 g·kg-1 merely. In terms of total potassium, the lowest contents were observed in the United States and Iran, with values of 4.00 and 3.10 g·kg-1, respectively. Besides, Iran showed the highest total nitrogen content of sludge, which was up to 62.90 g·kg-1. Probably, this is linked to the wastewater collection system. The better the collection system, the less the silt entering the sludge, so the higher the sludge organic matter content. Clearly, China still lagged behind developed countries like the United States, which needs to further improve its wastewater collection system.

Table 3 Content of organic matter and nutrients in sewage sludge from different countries and regions (g·kg^{-1})

Country and region (year *)	Organic matter	Total nitrogen	Total phosphorus	Total potassium	Literature
Jiujiang of China (2021-2022)	385.16	34.14	11.05	7.87	Present study
Shanxi of China (2005)	361.41-529.38	20.13-35.42	3.11-7.48	8.77-12.20	[12]
Guangxi of China (2014)	346.30	39.63	36.26	11.23	[13]
China (2003)	366.3	27.5	10.3	7.4	[14]
China (2008)	411.5	30.2	15.7	6.9	[15]
China (2009)	280	29.6	22.2	5.83	[16]
India (2014)	55.20-126.00	16.00-17.30	4.90-13.00	8.00-12.60	[17]
United States (2009)	534.00	26.00	8.10	4.00	[13]
Iran (2018)	336.00	62.90	14.10	3.10	[18]

* Year of sludge sampling.

3.2 Heavy metal contents of sewage sludge

In this study, heavy metals in sewage sludge from four typical WWTPs in Jiujiang City were tested for 12 consecutive months, and the test results were calculated for arithmetic average and standard deviation. Table 4 lists the computational results. On the whole, the sludge quality of the four WWTPs met the heavy metal content limit requirements as specified in the Chinese Quality of Sludge from Municipal Wastewater Treatment Plants (GB24188-2009), with average and maximum values all considerably less than the standard limits. Among them, the average Cu and Cr contents of sludge for WWTP A were far higher than the other three plants, which were up to 684.25 and 440.16 mg·kg-1, respectively, and were higher than the nationwide average values (see Table 5). Meanwhile, the average Zn contents of sludge for WWTPs C and D were higher than the other two plants, which exceeded 1,000 mg·kg-1, although were basically close to the nationwide average value.

From Table 5, it is clear that the average heavy metal contents of sewage sludge for the four WWTPs studied herein were all slightly higher than those for Shanxi [19] and Beijing of China[20] (except for the Ni content). Aside from the As content, the contents of other heavy metals were all less than or close to China's average values in 2003, 2008, and 2014. As the data suggests, with the standardization of the construction, management and supervision of centralized WWTPs in China's industrial parks, the heavy metal contents of sludge from municipal WWTPs has presented a declining trend, which is similar to the previous research conclusion [21]. Jiujiang City exhibited higher heavy metal contents of sewage sludge than Sweden, Iran, and Spain, but lower contents than Australia and India. The average metal concentrations of the samples in Turkey decreased as the following order of Zn > Cu > Cr > Pb > Ni > Mo > As > Cd > Se > Hg [7]， which is completely consistent with the order of the heavy metal contents in this study.

Table 4 Average content of heavy metals in sewage sludge and quality of sludge from municipal wastewater treatment plants (mg·kg^{-1})

WWTP	Statistics	Cd	Cu	Zn	Cr	Ni	Pb	Hg	As
A	Average *	2.02±0.20	684.25±59.38	582.33±50.18	440.16±99.66	34.62±9.18	28.67±9.78	0.50±0.15	30.24±5.03
	Range	1.54-2.47	514.56-787.04	468.02-701.46	105.66-586.17	16.78-68.92	13.55-60.49	0.16-0.95	16.14-47.48
B	Average *	1.87±0.23	121.03±9.42	462.10±42.86	83.29±29.46	42.40±4.87	29.82±13.09	0.84±0.17	29.18±10.26
	Range	1.44-2.81	103.43-159.63	365.53-640.55	33.21-200.86	30.48-58.27	5.18-60.78	0.26-1.16	12.36-62.39
C	Average *	1.33±0.21	313.37±29.88	1011.25±79.91	142.20±42.28	73.63±6.02	59.92±11.13	0.46±0.09	37.81±7.48
	Range	0.75-1.65	237.34-398.33	797.38-1175.08	62.03-253.53	51.24-92.31	38.19-105.35	0.30-0.76	22.35-55.37
D	Average *	2.16±0.24	102.33±14.19	1002.25±147.74	16.62±3.77	19.16±4.96	50.66±16.04	0.55±0.17	17.92±7.42
	Range	1.58-2.75	80.82-156.04	457.24-1342.87	8.07-27.17	10.92-42.61	10.91-110.43	0.17-0.92	7.54-53.3
Average concentration		1.85	305.25	764.48	170.57	42.45	42.27	0.59	28.79
GB24188-2009**		< 20	< 1500	< 4000	< 1000	< 200	< 1000	< 25	< 75

* Standard deviation: data size n = 12; confidence level P = 95%, t = 2.201.

*** Chinese Quality of Sludge from Municipal Wastewater Treatment Plants (GB24188-2009)*

Table 5 Content of heavy metals in sewage sludge from different countries and regions (mg·kg^{-1})

Country and region (year *)	Cd	Cu	Zn	Cr	Ni	Pb	Hg	As	Literature
Jiujiang of China (2021-2022)	1.85	305.25	764.48	170.57	42.45	42.27	0.59	28.79	Present study
Shanxi of China (2017)	0.65	100.53	331.5	147.72	34.28	11.59	-	11.56	[19]
Beijing of China (2016)	0.24	195.70	721.36	57.44	77.16	25.77	-	12.35	[20]
China (2014)	5.8	283.5	925.7	136.9	63.9	78.0	2.8	15.7	[21]
China (2008)	7.18	532.89	1270.37	221.99	79.11	114.99	3.80	16.94	[15]
China (2003)	2.97	185	1450	185	77.5	131	2.84	16.1	[22]
Sweden (2014)	0.7	314	498	30	15	16	0.5	-	[23]
Iran (2018)	-	10.3	94.3	5.6	3.4	7.4	0.14	1.6	[18]
India (2014)	32.3-154.5	186-330	161-2050	35.5-60	47.17-60	28.5-240	-	-	[17]
Australia (2011)	0.70-13.6	92-1996	210-3060	308	166	323	-	-	[17]

Spain (2009)	0.2-3.0	149-230	560-1100	1-210	25-71	43-219	-	-	[17]

* Year of sludge sampling.

Sludge disposal method varies among municipal WWTPs in China, and the standards on heavy metals differ as well (see Table 6). In an ascending order of content limits, these standards are the Control standards of pollutants in sludge for agricultural use, the Quality standards for sludge used in gardens or parks, the Quality standards for sludge used in land improvement, and the Quality standards of sludge for co-landfilling. The average values in sludge for the Jiangxi WWTPs studied herein were all in line with the above four standards. Nevertheless, the maximum values did not necessarily meet the standards: Cd (2.81), Cu (787.04), Zn (1342.87), Cr (586.17), Ni (92.31), Pb (110.43), Hg (1.16), and As (62.39). For instance, the maximum values of Cu, Zn, Cr, and As exceeded the limits specified by the Control standards of pollutants in sludge for agricultural use, while the maximum values of Cd and Ni were quite close to the Quality standards for sludge used in gardens or parks and the Quality standards for sludge used in land improvement. Hence, this study does not recommend the use of sewage sludge in Jiujiang City for agricultural use, and recommends its cautious use in gardens or parks and in land improvement. Nevertheless, the sludge can be definitely used for co-landfilling as a landfill cover material.

Table 6 Chinese criteria on heavy metals in sewage sludge from municipal wastewater treatment plants ($mg\cdot kg^{-1}$)

Chinese criteria	Cd	Cu	Zn	Cr	Ni	Pb	Hg	As
GB 4284-2018	< 3	< 500	< 1200	< 500	< 100	< 300	< 3	< 30
GB/T 23486-2009	<5	< 800	< 2000	< 600	< 100	< 300	< 5	< 75
GB/T 24600-2009	<5	< 800	< 2000	< 600	< 100	< 300	< 5	< 75
GB/T 23485-2009	< 20	< 1500	< 4000	< 1000	< 200	< 1000	< 25	< 75

GB 4284-2018：Control standards of pollutants in sludge for agricultural use

GB/T 23486-2009：Disposal of sludge from municipal wastewater treatment plants: Quality of sludge used in gardens or parks

GB/T 24600-2009：Disposal of sludge from municipal wastewater treatment plants: Quality of sludge used in land improvement

GB/T 23485-2009：Disposal of sludge from municipal wastewater treatment plants: Quality of sludge for co-landfilling

3.3 Feasibility analysis of sludge land application

3.3.1 Comparative analysis of sludge nutrients

Sewage sludge can be used as an important resource for agricultural utilization. The main reason is that it contains plenty of nutrients beneficial to plants and can be used as a soil improver to reduce the dependence on fertilizers. For Jiangxi Province, the organic matter content of sludge could be 633.28 g·kg-1 at maximum, and the average value could also reach 385.16 g·kg-1(see Table 7), which was very close to the average value (384 g·kg-1) of China's municipal sludge [24]. Comparison revealed that the organic matter content of sludge in Jiujiang City was 53.94%, 74.07% and 60.75% of pig manure, chicken manure and cow manure, respectively. In terms of nutrients, the sludge in Jiujiang City contained relatively high levels of nitrogen and phosphorus, as well as a relatively low level of potassium. Moreover, its total nutrient content reached 53.06 g·kg-1, which exceeded the total nutrient contents of pig, chicken and cow manures, although was lower than the total nutrient content (87.11 g·kg-1) of sludge in Guangxi Province [13]. Thus, the sludge in Jiujiang City conforms to Chinese Control standards of pollutants in sludge for agricultural use (GB 4284-2018), which stipulate that the organic matter content should be greater than 200 g·kg-1. Besides, it also meets the requirement that the total nutrient content of organic fertilizers in China should not be less than 40 g·kg-1 [25], which thus has good fertilizer efficiency and soil improving effect in agricultural production and gardening.

Table 7 Comparison of organic matter and nutrients in sewage sludge from Jiujiang City with pig manure, chicken manure and cow manure (g·kg^{-1})

Item	Organic matter	Total nitrogen	Total phosphorus	Total potassium	Total nutrient	Literature
Sewage sludge	385.16	34.14	11.05	7.87	53.06	Present study
Pig manure	714.00	20.70	9.00	11.20	40.90	[17]
Chicken manure	520.00	23.40	9.30	16.10	43.90	[17]
Cow manure	634.00	16.70	4.30	9.50	30.50	[17]

3.3.2 Analysis of soil pollution by sludge heavy metals

Heavy metals have always been the major factor hindering the agricultural application of sewage sludge. Extensive studies have been done concerning the soil pollution by heavy metals in sewage sludge. Currently, the Nemerow index method is the mainstream method for evaluating heavy metal pollution of soil. In this study, a combination of single-factor index method and Nemerow synthetic pollution index method was used to evaluate the heavy metal pollution caused by land application of sewage sludge. Tables 8 and 9 detail the relevant criteria for classification of pollution levels. Given the differing evaluation standards for heavy metal pollution adopted in extant studies, it is impossible to compare the evaluation results between many studies. For the present evaluation of heavy metal pollution of soil, the use of risk screening values for agricultural land pollution as specified in Chinese Soil Quality Control Criteria for Soil Pollution in Agricultural Lands (Trial) (GB 15618-2018) are recommended as the assessing standards. When the pollutants contents in agricultural soil are equal to or lower than these values, their risks to the quality and safety of agricultural products, the growth of crops or the soil ecological environment are low, which are negligible.

According to the evaluation index results in Table 10, various sludge heavy metals in Jiujiang City could be ranked as Cd> Cu> Zn> Cr> As> Ni> Pb> Hg in a descending order of single-factor pollution index. To be specific, Cd and Cu reached heavy pollution levels, Zn was at a medium pollution level, Cr was at a slight pollution level, while As, Ni, Pb, and Hg were at absolutely safe levels. For the four WWTPs, the Nemerow synthetic pollution indices of sludge heavy metals were A> D> C> B in a descending order. However, all of them reached heavy pollution levels. The high synthetic pollution levels were attributed to the larger single-factor pollution indices of Cd and Cu. Thus, Cd and Cu are the primary heavy metal pollution factors in the sewage sludge of Jiangxi, to which special attention should be given during selection of sludge disposal method.

Table 8 Classification criteria for heavy metal single pollution in soil

P_i	$P_i \leqslant 1.0$	$1.0 < P_i \leqslant 2.0$	$2.0 < P_i \leqslant 3.0$	$3.0 < P_i \leqslant 5.0$	$P_i > 5.0$
Pollution level	No pollution	Slight pollution	Mild pollution	Medium pollution	Heavy pollution

Table 9 Classification criteria for heavy metal Nemerow synthetic pollution in soil

P_N	$P_N \leqslant 0.7$	$0.7 < P_N \leqslant 1.0$	$1.0 < P_N \leqslant 2.0$	$2.0 < P_N \leqslant 3.0$	$P_N > 3.0$
Pollution level	No pollution	Warning line	Mild pollution	Medium pollution	Heavy pollution

Table 10 Pollution index of heavy metals and pollution level in soil

WWTP	Pi								PN	Pollution level
	Cd	Cu	Zn	Cr	Ni	Pb	Hg	As		

A	6.73	13.69	2.91	2.93	0.58	0.41	0.38	0.76	14.14	Heavy pollution
B	6.23	2.42	2.31	0.56	0.71	0.43	0.65	0.73	6.47	Heavy pollution
C	4.43	6.27	5.06	0.95	1.23	0.86	0.35	0.95	6.75	Heavy pollution
D	7.20	2.05	5.01	0.11	0.32	0.84	0.42	0.45	7.49	Heavy pollution
Average value	6.17	6.10	3.82	1.13	0.71	0.63	0.45	0.72	4.70	Heavy pollution
Pollution level	Heavy pollution	Heavy pollution	Medium pollution	Slight pollution	No pollution	No pollution	No pollution	No pollution		

3.3.3 Analysis of potential ecological risks of sludge heavy metals

Potential ecological risk index method is widely used in evaluating the heavy metal pollution of soil and sediment (see Table 11). In this study, the background values of soil environment in Jiujiang City were selected as the evaluation reference values, and Table 12 lists the relevant computational results. Respecting the potential ecological risk index of various heavy metals, Cd posed an extremely high ecological risk, Cu posed a medium ecological risk, while the remaining heavy metals had low ecological risks. In a descending order of potential ecological risk level, these heavy metals were Cd> Cu> As> Ni > Zn> Cr> Pb. For the four WWTPs, the synthetic potential ecological risk indices of heavy metals in sewage sludge all exceeded 480, all of which had extremely high ecological risks. Thus, clearly, Cd and Cu are the primary potential ecological risk factors in sewage sludge from Jiangxi Province, which pose rather high ecological risks in the case of land application. In particular, close attention should be paid to Cd. When using for soil improvement, the sewage sludge should be treated effectively or its use should be reduced, in order to lower the Cd content.

Table 11 Classification criteria for potential ecological risk level

Index	Potential ecological risk level				
C_f^i	$C_f^i<1$	$1\leqslant C_f^i<3$	$3\leqslant C_f^i<6$	$C_f^i\geqslant 6$	
E_r^i	$E_r^i<40$	$40\leqslant E_r^i<80$	$80\leqslant E_r^i<160$	$160\leqslant E_r^i<320$	$E_r^i\geqslant 320$
RI	$RI<120$	$120\leqslant RI<240$	$240\leqslant RI<480$	$RI\geqslant 480$	
Risk level	Slight	Medium	High	Very high	Extremely high

Table 12 Potential ecological risk level of heavy metal in soil

WWTP	E_r^i							RI	Risk level
	Cd	Cu	Zn	Cr	Ni	Pb	As		
A	606.00	164.48	8.44	18.34	9.11	4.47	29.07	839.91	Very high
B	561.00	29.09	6.70	3.47	11.16	4.64	28.06	644.12	Very high
C	399.00	75.33	14.66	5.93	19.38	9.33	36.36	559.99	Very high
D	648.00	24.60	14.53	0.69	5.04	7.89	17.23	717.98	Very high
Average value	555.00	73.38	11.08	7.11	11.17	6.58	27.68	692.00	Very high
Risk level	Extremely high	Medium	Slight	Slight	Slight	Slight	Slight		

3.3.4 Pondering on the land application of sewage sludge

Currently, the land application of sewage sludge is attributed more to the rich organic matter, nitrogen, phosphorus and other resources the sludge contains, so as to cope with the loss of nutrients (especially phosphorus) in the soil. Since phosphorus is a non-renewable resource, the European Union has introduced a series of phosphorus recycling policies. Targeting the sewage sludge, Germany promulgated the Amendment to Sludge Regulations (Draft) in 2016, with a goal of recovering phosphorus and other recyclable components in the sewage sludge. The amendment also restricts the traditional direct land application of sewage sludge. To recycle the phosphorus element in sewage sludge to agriculture without damaging the soil environment, the Swedish EPA has proposed the limit ratios of various heavy metals to phosphorus (mgMe2+·kgP-1), e.g. Cd (30), Cr (1200), Cu (17000), Zn (25000), Ni (1000), Pb (900), and Hg (20), with the aims of limiting the heavy metal contents and improving the sludge quality. The sludge can be used as the agricultural fertilizer if it reaches the concentration or ratio limit [23]. Clearly, higher requirements have been put forward on the sludge for agricultural use, which will have a profound impact on the future land application methods of sewage sludge. Given the dual attributes of sewage sludge as both environmental hazards and resources, its harmless disposal with advanced technologies is necessary first, after which its utilization as resources can be considered thoughtfully. Only by doing so, the economic and social benefits of sludge can be attained.

Since heavy metal pollution is one of the key factors in the land application of sewage sludge, it is very important and prudent to limit the contents of heavy metals in the sludge. Countries around the world have made detailed regulations on the limits of heavy metals in sludge for agricultural use (see Table 13). The heavy metal limits in the United States and France are rather high, which are far lower than the standards of other European countries. For China, the limits for heavy metal are lower than the United States and France. In the opinion of Harrison et al. [4], the current US federal regulations on the land application of sewage sludge fail to pay enough attention to the protection of agricultural production, ecological safety or human health, who recommended the implementation of more restrictive application standards. Sweden, despite being one of the EU countries with the strictest restrictions on heavy metals in sludge for agricultural use, has put forward stricter limits on such types of heavy metals, which will be implemented by 2030. Governments of various countries will impose ever stricter limits for heavy metals in sludge. This is also one aspect deserving our close attention in the future.

Table 13 Criteria of heavy metals in sludge for agricultural use in different countries ($mg·kg^{-1}$)

Country	Cd	Cu	Zn	Cr	Ni	Pb	Hg	As	Standard or literature
China	< 3	< 500	< 1200	< 500	< 100	< 300	< 3	< 30	GB 4284-2018
United States	< 39	< 1500	< 2800	-	< 420	< 300	< 17	< 41	[4]
Denmark	< 0.8	< 1000	< 4000	< 100	< 30	< 120	< 0.8	-	[23]
France	< 20	< 1000	< 3000	< 1000	< 200	< 800	< 10	-	[23]
Sweden	< 2	< 600	< 800	< 100	< 50	< 100	< 2.5	-	[23]
Proposed legislation 2030, Sweden	< 0.8	< 475	< 700	< 35	< 30	< 25	< 0.6	-	[23]

Although mandatory limits have been set for heavy metal contents, studies from various countries have still found that the land application of sewage sludge can alter the physical, chemical and biological properties of soil, and even lead to soil ecological issues and food chain pollution. According to a study by Kelly et al. [26], the number of culturable bacteria in sludge-improved soil increased significantly, while the dehydrogenase activity decreased by 20 times. Besides, pronounced changes were noted in the microbial community structure, with an increase of resistance to zinc by nearly 20 times. Shen et al. [27] argued that many projects have exceeded the requirements of secondary soil standards according to the increased pollutant contents caused by continuous sludge application, and that the implementation of existing agricultural sludge standards would result in significantly deteriorated quality of soil environment. A study on the impact of sewage sludge on soil by Wang et al. [28] found that the Pb and Cu exceeded the grade A standards of soil environment, although did not exceed the grade B standards. Besides, the Cd exceeded the grade B standard. They recommended not to apply the sewage sludge to farmlands with a grade B or above soil quality. According to another study, the Hg, Zn, and Cu contents in the surface soil of urban sludge application areas in Beijing increased by 3731%, 86.3%, and 63.0%, respectively, showing a rather apparent cumulative trend, indicating that the land application of municipal sludge would cause greater risk of Cu pollution [29]. Through field experiments, Sun et al. [30] discovered that the Cd and Pb contents in surface soil increased markedly, and the Cr and Pb contents in partial wheat grains from sludge treatment exceeded the national standards on food hygiene. Nevertheless, antibiotics, drug-resistant bacteria and genes in the sewage sludge could be released into the environment via land application, so further evaluation was necessary regarding the risk of land application of sewage sludge to the human and environmental health [31]. Hence, it is necessary to strengthen the long-term monitoring of land application of sewage sludge, and carry out research focusing on the accumulation, migration and transformation of pollutants and the assessment of their risks to the ecological environment and human health.

4. Conclusion

In this paper, the components of sewage sludge from four typical WWTPs in Jiujiang City of China are monitored for one consecutive year, and the feasibility of its land application is evaluated and analyzed on the basis of the test results. The following conclusions are drawn:

(1) With average contents of organic matter, total nitrogen, total phosphorus, total potassium, and total nutrients being 385.16, 34.14, 11.05, 7.87, and 53.06 $g{\cdot}kg^{-1}$, respectively, the studied sewage sludge conforms to Chinese Control Standards of Pollutants in Sludge for Agricultural Use (GB 4284-2018), which stipulate that the organic matter content should be greater than 200 $g{\cdot}kg^{-1}$. Besides, it also meets the requirement that the total nutrient content of organic fertilizers in China should not be less than 40 $g{\cdot}kg^{-1}$, which thus has good land application value in agricultural production and landscaping.

(2) Average contents of Cd, Cu, Zn, Cr, Ni, Pb, Hg and As in the sewage sludge are 1.85, 305.25, 764.48, 170.57, 42.45, 42.27, 0.59 and $mg{\cdot}kg^{-1}$, respectively, which are less than or close to China's average values (except for As), thus conforming to the limits for heavy metal contents as specified in the Quality of Sludge from Municipal Wastewater Treatment Plants (GB24188-2009). However, the maximum values of Cu, Zn, Cr and As exceed the limits of pollutant control standards on sludge for agricultural use; and the maximum values of Cu and Cr are also close to the limits of quality standards for sludge used in gardens or parks. Hence, the sewage sludge in Jiujiang City is unsuitable for use in agricultural production, and should be used with caution in landscaping and land improvement. Nevertheless, the sludge can be used for co-landfilling as a cover material.

(3) In a descending order of single-factor pollution index, various heavy metals are ranked as follows: Cd> Cu> Zn> Cr> As> Ni> Pb> Hg. Among them, Cd and Cu reach heavy pollution levels, Zn is at a medium pollution level, Cr is at a slight pollution level, while As, Ni, Pb, and Hg are at absolutely safe levels.

(4) In a descending order of potential ecological risk level to soil, various heavy metals are ranked as follows: Cd> Cu> As> Ni> Zn> Cr> Pb. Among them, Cd poses an extremely high ecological risk, Cu poses a medium ecological risk, while the remaining heavy metals have low ecological risks.

(5) Ever stricter limits will be imposed on the heavy metals in sewage sludge, which will have a profound impact on the future land application methods of the sludge. In the future, long-term monitoring of sewage sludge land application should be strengthened, and the research emphases

should be on the accumulation, migration and transformation of pollutants and the assessment of their risks to the ecological environment and human health.

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References

[1] Ma P, Rosen C. Land application of sewage sludge incinerator ash for phosphorus recovery: A review[J]. Chemosphere, 2021, 274: 129609

[2] Bagheri M, Bauer T, Burgman L E, et al. Fifty years of sewage sludge management research: Mapping researchers' motivations and concerns[J]. Journal of Environmental Management, 2023, 325: 116412.

[3] Collivignarelli M C, Abbà A, Frattarola A, et al. Legislation for the Reuse of Biosolids on Agricultural Land in Europe: Overview[J]. Sustainability, 2019, 11: 6015

[4] Lofty J, Muhawenimana V, Wilson C a M E, et al. Microplastics removal from a primary settler tank in a wastewater treatment plant and estimations of contamination onto European agricultural land via sewage sludge recycling[J]. Environmental Pollution, 2022, 304: 119198.

[5] Chojnacka K, Skrzypczak D, Szopa D, et al. Management of biological sewage sludge: Fertilizer nitrogen recovery as the solution to fertilizer crisis[J]. Journal of Environmental Management, 2023, 326: 116602.

[6] Kanteraki A E, Isari E A, Svarnas P, et al. Biosolids: The Trojan horse or the beautiful Helen for soil fertilization?[J]. Science of The Total Environment, 2022, 839: 156270.

[7] Yakamercan E, Ari A, Aygün A. Land application of municipal sewage sludge: Human health risk assessment of heavy metals[J]. Journal of Cleaner Production, 2021, 319: 128568

[8] Gomes L A, Gabriel N, Gando-Ferreira L M， et al. Analysis of potentially toxic metal constraints to apply sewage sludge in Portuguese agricultural soils[J]. Environmental Science and Pollution Research, 2019, 26(25): 26000-26014.

[9] He Jie, Zhang Xu, Xiao Dan, et al. Feasibility of land application of sewage sludge from Kunming wastewater treatment plants[J]. China Water & Wastewater, 2010, 26(17): 103-105. (in Chinese)

[10] Chu L, He W, Xu F, et al. Ecological risk assessment of toxic metal(loid)s for land application of sewage sludge in China[J]. Science of The Total Environment, 2022, 836: 155549.

[11] Hakanson L. An ecological risk index for aquatic pollution control. a sedimentological approach[J]. Water Research, 1980, 14(8): 975-1001.

[12] Sun Xining, Zhang Zengqiang, Zhang Yongtao, et al. Components change of sewage sludge by month and land application in Shaanxi Yangling Sewage Treatment Plant[J]. Journal of Northwest A & F University (Nature Science Edition), 2007, 35(9): 215-220. (in Chinese)

[13] Zhang Jun, Zhang Zhengshi, Wang Dunqiu, et al. Physicochemical property and potential ecological risk assessment of heavy metals in sludge from sewage plants in Guangxi[J]. Environmental Engineering, 2014, 32(1): 108-112. (in Chinese)

[14] National Bureau of Statistics, Ministry of Environmental Protection. China environmental statistics yearbook in 2008[M]. Beijing: China Statistics Press，2009. (in Chinese)

[15] Ma Xuewen, Weng Huanxin, Zhang Jinjun. Regional characteristics and trend of heavy metals and nutrients of sewage sludge in China[J]. China Environmental Science, 2011, 31(8): 1306-1313. (in Chinese)

[16] Guo Guanghui, Yang Jun, Chen Tongbin. Concentrations and variation of organic matter and nutrients in municipal sludge of China[J]. China Water & Wastewater, 2009, 25(13): 120-121. (in Chinese)

[17] Sharma B, Sarkar A, Singh P, et al. Agricultural utilization of biosolids: A review on potential effects on soil and plant grown[J]. Waste Management, 2017, 64: 117-132.

[18] Galangash M M, Mahdavianpour M, Safa S G. Characterization of an Industrial Sewage Sludge and Its Evaluation for Land Application[J]. Iranian Journal of Toxicology, 2018, 12(5): 27-34.

[19] Duan B, Zhang W, Zheng H, et al. Disposal situation of sewage sludge from municipal wastewater treatment plants (WWTPs) and assessment of the ecological risk of heavy metals for its land use in Shanxi, China[J]. International Journal of Environmental Research and Public Health, 2017, 14: 823.

[20] Meng Guoxin, Zha Tonggang, Zhang Xiaoxia, et al. Heavy metal pollution characteristics and ecological risk assessment of the sludge from wastewater treatment plants in Beijing[J]. Ecology and Environmental Sciences, 2017, 26(9): 1570-1576. (in Chinese)

[21] Guo Guanghui, Chen Tongbin, Yang Jun, et al. Regional distribution characteristics and variation of heavy metals in sewage sludge of China[J]. Acta Scientiae Circumstantiae, 2014, 34(10): 2455-2461. (in Chinese)

[22] Chen Tongbin, Huang Qifei, Gao Ding, et al. Heavy metal concentrations and their decreasing trends in sewage sludges of China[J]. Acta Scientiae Circumstantiae, 2003, 23(5): 561-569. (in Chinese)

[23] Mattsson A, Finnson A, I'Ons D. Heavy metal content of Swedish municipal wastewater sludge- status and goals[J]. Water Science & Technology, 2017, 76(4): 869-876.

[24] Li Yanxia, Chen Tongbin, Luo Wei, et al. Contents of organic matter and major nutrients and the ecological effect related to land application of sewage sludge in China[J]. Acta Ecologica Sinica, 2003, 23(11): 2464-2473. (in Chinese)

[25] National Agricultural Technology Extension Center. Chinese Organic Fertilizers Nutrients[M]. Beijing: China Agricultural Press, 1999. (in Chinese)

[26] Kelly J J, Haggblom M, Tate III R L, et al. Effects of the land application of sewage sludge on soil heavy metal concentrations and soil microbial communities[J]. Soil Biology and Biochemistry, 1999, 31: 1467-1470.

[27] Shen Rongyan, Luo Yongming, Teng Ying, et al. The advances in municipal sewage sludge and assessment for its land application[J]. Soil, 2006, 38(5): 517-524. (in Chinese)

[28] Wang X, Chen T, Ge Y, et al. Studies on land application of sewage sludge and its limiting factors[J]. Journal of Hazardous Materials, 2008, 160(2-3): 554-558.

[29] Li Yabin, Yang Jun, Lei Mei, et al. Pollution risk assessment of heavy metals in sewage sludge for land application in Beijing[J]. China Water & Wastewater, 2015, 31(9): 117-120. (in Chinese)

[30] Sun Na, Shang Heping, Ru Shuhua, et al. Effects of continuous application of sewage sludge compost on heavy metals accumulation and mobility characteristics in soil profile and on heavy metals uptake of wheat[J]. Environmental Science, 2017, 38(2): 815-824. (in Chinese)

[31] Bondarczuk K, Markowicz A, Piotrowska-Seget Z. The urgent need for risk assessment on the antibiotic resistance spread via sewage sludge land application[J]. Environment International, 2016, 87: 49-55.