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Classification and Disposal Strategy of Excess Sludge in the Petrochemical Industry

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Abstract: The excess sludge in the petrochemical industry is large in quantity, complex in composition, and highly harmful, and its rational disposal is of great significance for environmental protection and sustainable development. In the present study, a classification and disposal strategy for the excess sludge in the petrochemical industry is proposed. The strategy first analyzes the dioxin, flammability, corrosivity, reactivity, and leaching properties of the sludge, from which the waste type of the sludge (general waste or hazardous waste) can be determined. Then, methods of disposal can be selected depending on the type of waste and the corresponding risk analysis, enabling rationalized disposal of the sludge. To verify the effectiveness and practicability of the proposed sludge classification and disposal strategy, research on petrochemical excess sludge samples (i.e., Ah, Bl, and Cq) originated from three different regions in China is carried out as a case study. The component analysis of the above three sludge samples revealed that they are all general wastes. In addition, the possibility of employing Cq sludge for landfill, soil modification, and greening mud, as well as the risk of landfill and incineration disposal in solid waste landfills are investigated. Furthermore, natural radioactive elements uranium and thorium in Cq sludge sample are studied. The results show that Cq sludge cannot be used for landfill, soil modification, and greening mud due to excessive arsenic content. The proposed strategy provides a basis for the selection of reasonable petrochemical excess sludge disposal methods.

Keywords: excess sludge; disposal strategy; radioactive elements; petrochemical industry

1. Introduction

With the strengthening of public crisis awareness, governments have adopted stricter environmental protection policies and increased environmental supervision and punishment. The amount of petrochemical excess sludge is increasing year by year with the increase in demand for petrochemicals, and the potential environmental risks are increasing due to the particularity of the industry. The rational disposal of petrochemical excess sludge is of great significance to public health and environmental protection. The excess sludge has the characteristics of high water content, large quantity, and complex pollution components. Therefore, reasonable disposal methods of the remaining sludge should be dependent on its pollutant composition analysis and hazard research.

At present, various approaches for excess sludge pollutants treatment, such as harmless treatment, sludge utilization, pyrolysis [1,2], and incineration [3–5], have been widely utilized. Harmless

treatment involves bioleaching of heavy metals from sewage sludge [6] and biochemical treatment of sewage sludge [7], etc. Sludge utilization involves preparation of biochar [8] and adsorbent [9], recovery of phosphorus from excess sludge [10], methane production by anaerobic digestion of excess sludge [11,12], and additive of the cement [13], etc. For example, Wu et al. [14] adopted the biodegradable chelating agent N,N-bis(carboxymethyl) glutamic acid to remove harmful substances, such as cadmium, nickel, and copper, in sludge. Choi et al. [12] utilized excess sludge to produce methane via anaerobic digestion after hydrothermal pretreatment of sludge at 180 °C for 76 min. Alvarez et al. [15] employed conical spouted bed reactor to transform excess sludge into bio-oil at the temperature ranging from 450 to 600 °C. Moško et al. [3] used a fluidized bed to investigate residual sludge incineration pollutant emission reduction process. Although a large variety of reports on the disposal of excess sludge, the environmental risks arising from the disposal of excess sludge need to be evaluated. For example, with respect to the excess sludge for land using, the potential for soil improvement and utilization [16] and the impact on plants [17,18] should be analyzed. Meanwhile, the migration of heavy metals of excess sludge in the process of soil utilization should be studied [19]. With respect to thermal treatment of the excess sludge, it is necessary to evaluate the production of environmentally hazardous substances such as Polychlorinated dibenzodioxin and Polychlorinated dibenzofuran (PCDD/Fs) [20], hydrocarbons (PAHs) [21], and particulate matter [22] during sludge pyrolysis or incineration. Additionally, the migration [23,24] or removal [25] of heavy metals from the excess sludge should be studied.

Furthermore, some review studies are listed to summarize the existing technologies for disposal of sludges. For example, Qian et al. [26] summarized the treatment of sludge in supercritical water avoiding the pre-drying procedure and secondary pollution, and analyzed supercritical water gasification, supercritical water partial oxidation and supercritical water oxidation technologies of sludge. Recent studies showed that through vermicomposting, industrial sludge could be transformed into matured organic fertilizer or vermicompost in a shorter period. To this end, Lee et al. [27] analyzed the utilization of vermicomposting process to manage industrial sludge in order to assess the feasibility of the technology. In order to improve the energy yield, Liu et al. [28] evaluated the anaerobic fermentation process of sludge which was capable of obtaining bioenergy by biogas production under the functions of microbes. For the purpose of obtaining a final product of sludge which can be used as fertilizers in farm soils, Pereira et al. [29] discussed removal technologies of toxic metals from sewage sludge through chemical, physical, and biological treatments. Additionally, review studies on thermal hydrolysis [30], nitrogen, and phosphorus removal [31] of sludges are well presented.

Despite the large variety of approaches that have been established on the disposal of excess sludges, most of the technologies are still in the theoretical exploration stage with very high cost for industrial application. Therefore, the sludge disposal method must be rationally selected [32].

The excess sludge in the petrochemical industry is produced by the sewage treatment of petroleum refining and chemical production processes. It has typical industrial characteristics. Therefore, the choice of the sludge disposal method in the petrochemical industry cannot be completely based on the disposal method of municipal sludge. Physical and chemical properties of the excess sludge should be studied in depth so as to select effective disposal methods, and to establish reasonable sludge disposal strategy, for providing reference on disposal of excess sludge in the petrochemical industry. However, the research on the pollution components and dangers of excess sludge in the petrochemical industry has not been reported publicly. In addition, the basic data and reference basis for the research on the targeted disposal of excess sludge in the petrochemical industry are lacking. Furthermore, due to the absence of comprehensive disposal strategies on petrochemical excess sludge, various disposal methods are improperly selected and adopted, giving rise to a series of environmental issues.

In this study, a novel classification and disposal strategy for petrochemical excess sludge is proposed. Based on the strategy, hazard analysis of petrochemical excess sludge samples obtained from three different Chinese regions are performed. In addition, environmental impacts caused by different disposal approaches of the Cq sludge sample are analyzed. Specifically, as natural radioactive

elements that are widespread in the Earth's crust, uranium and thorium in the Cq sludge sample are investigated. To the best of our knowledge, no previous published work was found in which the analysis of radioactive elements in petrochemical excess sludge have been involved. The proposed strategy provides a basis for the selection of reasonable petrochemical excess sludge disposal methods.

2. Experimental Section

2.1. Materials

Ah, Bl, and Cq sludge were taken from three refining and chemical enterprises Ah (South area, China), Bl (Central Shandong Province, China), and Cq (Eastern Shandong Province, China), respectively. After sampling, they were centrifuged to a moisture content of about 85%.

2.2. Analytical Methods

Leaching of the volatile organic compounds in sludge: 110 g of sludge was added into a zero-headspace extraction vessel (ZHE, Anyi Co., Ltd., Anhui, China). Then, deionized water was added into the sludge to ensure the liquid-solid ratio (L: Kg) of 10:1. The ZHE was then fixed in an inverted oscillator (AYZ-K, Anyi Co., Ltd., Anhui, China) and shook at 30 r/min for 19 h. After the oscillation, the leachate was collected and stored under cold conditions [33].

Leaching of the heavy metals and the non-volatile organic compounds in sludge: 180 g of sludge was placed in a Teflon bottle. Then, an acid solution with a pH of 3.2 (configured by a mixture of concentrated sulfuric acid and concentrated nitric acid with a mass ratio of 2:1) was added into the Teflon bottle to maintain the liquid-solid ratio (L:kg) of 10:1. The bottle was then fixed in a flip-type shaker and shaken at a speed of 30 r/min for 19 h. After the oscillation, the leachate was collected and stored under cold conditions.

High-temperature incineration of sludge: 500 g of dried sludge was placed in a high-temperature muffle furnace (QSX-3-12, Cinite Co., Ltd., Hebei, China) and heated to 850 °C for 10 min. After incineration, the ash was taken out and cooled to room temperature.

The volatile organic compounds in the leachate were all determined by a gas chromatography-mass spectrometry (GC-MS) (7890b-5977b, Agilent Co., Ltd., Palo Alto, CA, USA), except for the ethylbenzene which was determined by a gas chromatography (GC) (7890B, Agilent Co., Ltd., Palo Alto, CA, USA). The test conditions were in accordance with Chinese Environmental Protection Industry Standard HJ/299-2007 [34].

The non-volatile organic compounds in the leachate including dinitrobenzene, phenol, 2,4-dichlorophenol, 2,4,6-trichlorophenol, benzopyrene, and dibutyl phthalate were determined by GC-MS; the nitrobenzene was determined by a high-performance liquid chromatography (HPLC) (1260VWD, Agilent Co., Ltd., Palo Alto, CA, USA); p-nitrchlorobenzene, 2,4-dinitrochlorobenzene, pentachlorophenol, and dioctyl phthalate were determined by high-performance liquid chromatography–mass spectrometry (HPLC-MS) (Agilent 1100MSD, Agilent Co., Ltd., Palo Alto, CA, USA); the polychlorinated biphenyl was determined by GC. The test conditions were in accordance with Chinese Environmental Protection Industry Standard HJ/299-2007 [34].

The metal elements in the leachate, sludge and ash were measured by an inductively-coupled plasma mass spectrometer (ICP-MS) (ICP-6300, Thermo Sentific Co., Ltd., Waltham, MA, USA).

The dioxin contents in the sludge were determined by a high-resolution gas chromatography high-resolution mass spectrometry (HRGC-HRMS) (Agilent 6890 GC/Waters AutoSpec Premier, USA) in accordance with Chinese Environmental Protection Industry Standard HJ/77.3-2008 [35]. The petroleum hydrocarbon contents in the sludge were measured by an infrared spectrometer (Nicolet 6700, Thermo Fisher Scientific Co., Ltd., Massachusetts, USA) in accordance with Chinese National Standard 5085.6-2007 [36]. The contents of C, H, and N were determined in accordance with the method specified in Chinese National Standard GB/T 30733-2014 [37]; the content of O was determined in accordance with the method specified in Chinese National Standard GB/T 476-2001 [38]; the contents

of S, Cl, and P were determined in accordance with the methods specified in Chinese National Standard GB/T 214-2007 [39], British Standard—European Norm BS EN 14582-2016 [40], and Chinese Environmental Protection Industry Standard HJ 712-2014 [41], respectively.

The corrosiveness, flammability, and reactivity of petrochemical excess sludge samples were tested in accordance with Chinese National Standards GB 5085.1-2007 [42], GB 5085.4-2007 [43], and GB 5085.5-2007 [44], respectively.

2.3. Strategy Description

In this work, a novel classification and disposal strategy for petrochemical excess sludge is proposed, as shown in Figure 1:

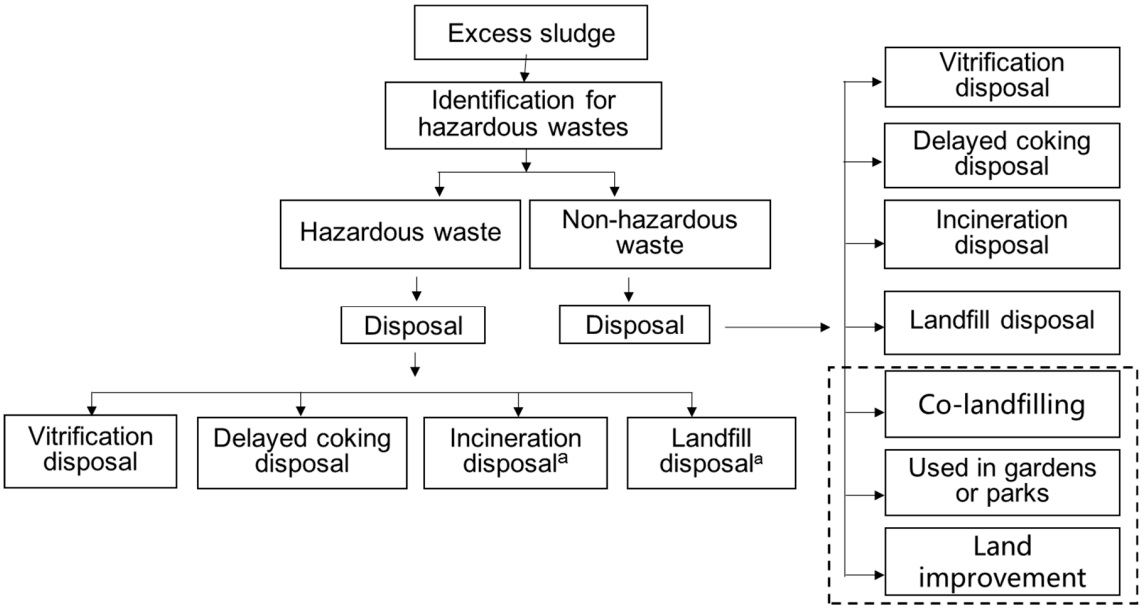

Figure 1. Flow chart of the classification and disposal strategy for petrochemical excess sludge. ([a] Meets the corresponding disposal requirements for hazardous waste.)

Firstly, the excess sludge should be identified. The identification method was in accordance with the Chinese National Standard GB/T 5085 [34,36,42–46]. The residual sludge identification in the petrochemical industry mainly examined the leaching toxicity, flammability, corrosivity, reactivity, and dioxin content of the inorganic and organic matter in the excess sludge. If the identification result shows that the sludge is hazardous waste, the vitrification treatment should be considered, since the high-temperature melting environment can completely decompose the organic components, and the inorganic components are fixed in the vitreous body to achieve complete harmlessness. Secondly, if there is a delayed cooker, the sludge disposed by the delayed cooker may be considered, but petroleum coke products should meet the corresponding product grade requirements specified in the Chinese Petrochemical Industry Standard (NB/SH/T 0527-2015) [47]. The hazardous waste incineration disposal method can be selected again, and the disposal process must meet the requirements of Chinese National Standard (GB 18484-2001) [48]. Finally, in the case of hazardous waste landfills with sufficient capacity, the remaining sludge can be disposed of in hazardous waste landfills after drying and reduction. Nevertheless, landfill disposal is not recommended, as it may be necessary to dig for re-disposal with changes in regional planning.

If the result of the identification show that the sludge is general solid waste, from the point of view of complete harmlessness, vitrification disposal should be preferentially selected. Likewise, coking disposal can be considered. If we consider the choice of general solid waste incineration disposal, the incineration temperature should be controlled above 850 °C [49]. We can also consider the landfill

of general solid waste landfill. If the excess sludge produced by the combined treatment of sewage and urban sewage can be tested for the possibility of using landfill mixed landfill, land improvement, or greening mud, it must meet the requirements of Chinese Standards GB/T 23485-2009 [50], GB/T 24600-2009 [51], and GB/T 23486-2009 [52], respectively.

For the selection of a reasonable disposal method, it is necessary to combine the risk analysis of the corresponding disposal method.

3. Results and Discussions

3.1. Judgment of Hazardous Solid Waste Properties of Petrochemical Excess Sludge

3.1.1. Risk Analysis of Petrochemical Excess Sludge Leaching

Leaching Toxicity of Heavy Metal Elements in Petrochemical Excess Sludge

The determined values of inorganic elements in the leachate after the leaching experiments of Ah, Bl and Cq wet sludge having a water content of 85% are shown in Table 1. It can be seen from Table 1 that the measured values of Cu, Zn, Pb, Cd, Be, Ni, Ag, Ba, Hg, As, and Cr^{+6} of Ah, Bl, and Cq wet sludge are far below the limit value, indicating that the Ah, Bl, and Cq sludge samples do not have the leaching toxicity of these inorganic elements.

Table 1. Determination of inorganic elements in excess sludge leachate.

Element	Leaching Concentration (mg/L)			Limit Value [34] (mg/L)
	Ah	Bl	Cq	
Cu	0.0348	0.0081	0.0130	100
Zn	0.162	0.080	1.020	100
Pb	0.009	0.004	0.003	5
Cd	0.00006	0.0011	0.0008	1
Be	N.D.	N.D.	0.0002	0.02
Ni	0.0867	0.0160	0.1021	5
Ag	0.0032	0.0005	0.0016	5
Ba	0.066	0.339	2.618	100
Cr^{+6}	0.004	N.D.	N.D.	5
Hg	N.D.	N.D.	N.D.	0.1
As	0.0011	0.0009	0.0026	5

Note: N.D.: not detected, below detection limit.

Leaching Toxicity Analysis of Organic Matter in Petrochemical Excess Sludge

The measured values of non-volatile and volatile organic compounds in the petrochemical excess sludge leachate are shown in Tables 2 and 3, respectively. It can be seen from Tables 2 and 3 that the non-volatile organic compounds and volatile organic compounds in the leaching solution of Ah, Bl, and Cq excess sludge samples were not detected except for the dibutyl phthalate and carbon tetrachloride of the Bl sample. It was detected that the content of non-volatile organic matter and volatile organic compounds in all sample leachates were below the limit.

From analysis of leaching toxicity of inorganic elements and organic compounds in petrochemical excess sludge, it was found that all the measured substances did not exceed their limit values. This may be attributed to the current management measures for sewage integration in various refining and chemical enterprises. The sewage that is incorporated into the sewage treatment plant needs to be evaluated and analyzed in advance, in which those with high toxic content are not allowed to be incorporated into the sewage treatment plant pipe network and needs to be isolated for specific disposal.

Table 2. Determination of non-volatile organic matter in excess sludge leachate.

Name	Leaching Concentration (mg/L)			Limit Value [34] (mg/L)
	Ah	Bl	Cq	
Nitrobenzene	N.D.	N.D.	N.D.	20
Dinitrobenzene	N.D.	N.D.	N.D.	20
p-Nitrchlorobenzene	N.D.	N.D.	N.D.	5
2,4-Dinitrochlorobenzene	N.D.	N.D.	N.D.	5
Pentachlorophenol	N.D.	N.D.	N.D.	50
Phenol	N.D.	N.D.	N.D.	3
2,4-Dichlorophenol	N.D.	N.D.	N.D.	6
2,4,6-Trichlorophenol	N.D.	N.D.	N.D.	6
Benzopyrene	N.D.	N.D.	N.D.	<0.0003
Dibutyl phthalate	N.D.	0.063	N.D.	2
Dioctyl phthalate	N.D.	N.D.	N.D.	3
Polychlorinated biphenyl	N.D.	N.D.	N.D.	0.002

Table 3. Determination of volatile organic compounds in excess sludge leachate.

Name	Leaching Concentration (mg/L)			Limit Value [34] (mg/L)
	Ah	Bl	Cq	
Benzene	N.D.	N.D.	N.D.	1
Toluene	N.D.	N.D.	N.D.	1
Ethylbenzene	N.D.	N.D.	N.D.	4
Xylene	N.D.	N.D.	N.D.	4
Chlorobenzene	N.D.	N.D.	N.D.	2
1,2-Dichlorobenzene	N.D.	N.D.	N.D.	4
1,4-Dichlorobenzene	N.D.	N.D.	N.D.	4
Acrylonitrile	N.D.	N.D.	N.D.	20
Trichloromethane	N.D.	N.D.	N.D.	3
Carbon tetrachloride	N.D.	0.067	N.D.	0.3
Trichloroethylene	N.D.	N.D.	N.D.	3
Carbon tetrachloride	N.D.	N.D.	N.D.	1

3.1.2. Analysis of Flammability, Corrosivity and Reactivity of Petrochemical Excess Sludge

The petrochemical excess sludge samples were tested for corrosiveness, flammability, and reactivity according to the requirements of Chinese National standards GB 5085.1-2007 [42], GB 5085.4-2007 [43], and GB 5085.5-2007 [44], respectively. The results of the sample test showed that the samples were not reactive, flammable, and corrosive.

3.1.3. Analysis of Dioxins in Petrochemical Excess Sludge

The contents of dioxins in Ah, Bl, and Cq wet sludge with a water content of 85% are shown in the Table 4. It can be seen from Table 4 that the dioxin substances in Ah, Bl, and Cq wet sludge were not detected except for octachlorinated dibenzofurans (OCDF) and octachlorinated dibenzo-p-dioxins (OCDD). The international toxic equivalence quantity (I-TEQ) values of Ah, Bl, and Cq wet sludge dioxin were 0.20, 0.36 and 0.27 ng-TEQ/kg, respectively. The I-TEQ values of PCDD/Fs in Ah, Bl, and Cq wet sludge is lower than the in Ah, Bl, and Cq wet sludge is lower than the dioxin content in the identification of hazardous wastes in GB 5085.6-2007 (15 μg-TEQ/kg) [36]. Similar results are found in the studies of sewage sludge from Dai et al. [20] that OCDD and OCDF also dominate the total PCDD/Fs amount of wet sewage sludge, and the I-TEQ value of PCDD/Fs is below the limit value specified in Chinese national Standard GB 5085.6-2007 [36].

Table 4. Dioxin content in wet sludge of Ah, Bl, and Cq.

Dioxins		I-TEF [35]	Measured Concentration (ng/kg)			I-TEQ (ng-TEQ/kg)		
			Ah	Bl	Cq	Ah	Bl	Cq
Polychlorinated dibenzofurans (PCDFs)	2,3,7,8-TCDF	×0.1	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	1,2,3,7,8-PeCDF	×0.05	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	2,3,4,7,8-PeCDF	×0.5	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	1,2,3,4,7,8-HxCDF	×0.1	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	1,2,3,6,7,8-HxCDF	×0.1	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	2,3,4,6,7,8-HxCDF	×0.1	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	1,2,3,7,8,9-HxCDF	×0.1	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	1,2,3,4,6,7,8-HpCDF	×0.01	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	1,2,3,4,7,8,9-HpCDF	×0.01	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	OCDF	×0.001	39.56	172.31	157.44	0.04	0.17	0.16
Polychlorinated dibenzodioxins (PCDDs)	2,3,7,8-TCDD	×1	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	1,2,3,7,8-PeCDD	×0.5	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	1,2,3,4,7,8-HxCDD	×0.1	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	1,2,3,6,7,8-HxCDD	×0.1	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	1,2,3,7,8,9-HxCDD	×0.1	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	1,2,3,4,6,7,8-HpCDD	×0.01	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
	OCDD	×0.001	158.31	192.05	106.83	0.16	0.19	0.11
Total dioxin (PCDD/Fs)		—	—	—	—	0.20	0.36	0.27

Note: I-TEF: international toxic equivalence factor; I-TEQ: international toxic equivalence quantity. N.D.: not detected, below detection limit (detection limit >0.01 ng/kg).

The leaching toxicity of Ah, Bl and Cq wet sludges and their flammability, corrosivity, reactivity and dioxin content were evaluated. The result showed that Ah, Bl, and Cq wet sludges are not hazardous wastes according to Chinese National Standard GB 5085.7-2007 [46].

3.2. Comprehensive Analysis of Risk of Petrochemical Excess Sludge

The results of the leaching toxicity of Ah, Bl, and Cq wet sludge and their flammability, corrosivity, reactivity and dioxin content were evaluated. It is known that Ah, Bl, and Cq wet sludge are not hazardous wastes.

3.3. Analysis of Petrochemical Excess Sludge Disposal

From Section 3.1, Ah, Bl, and Cq wet sludge are not hazardous waste. Hence, there are many ways to dispose of petrochemical excess sludge. Since the petrochemical enterprise Cq mainly uses inferior crude oil as raw materials for production, the comprehensive analysis of Cq sample is considered more representative. Therefore, we selected Cq sludge sample as an example to analyze the risks of landfill, land improvement, greening mud, and incineration disposal.

Analysis of Mixed Landfill, Land Improvement and Greening Mud for Petrochemical Excess Sludge Landfill

In this study, Cq sludge was taken as an example to compare the element content of Cq sludge with the mixed landfill, land improvement and greening mud of the sludge dump. The mixed landfill and land improvement of the sludge dump, as well as the possibility of greening mud were analyzed.

The content of Cq sludge sample elements is shown in Table 5. It can be seen from Table 5 that the content of As element exceeds the standard according to the standard regulations for mixed landfill, land improvement and greening sludge for sludge dumps. Therefore, it is not possible to use the sludge for mixed landfill, land use and mud for greening. In addition, the content of Al in the excess sludge is relatively high, reaching 6434.88 mg/L. The aluminum in the excess sludge is mainly from the polymerized aluminum flocculant in the sewage treatment plant. The polymerized aluminum flocculant is commonly adopted in sewage treatment plant of the refining and chemical industry. In areas where acid rain is frequent and hence the soil pH is low, aluminum is easily soluble in surface water, threatening severely to animals and plants. Noticing that the safety threshold of aluminum content in tap water is 0.2 mg/L. Aluminum can bind to proteins, and can also bind to lipids, sugars and nucleic acids, etc., interfering with some ion metabolism in plant cells, and affecting various physiological and biochemical processes. Additionally, the existing of aluminum inhibits plant root tip cell elongation and cell division, and affects the biosynthesis of plant cell walls, thereby inhibiting the growth of plants. The perniciousness of aluminum to human health is mainly reflected in the aluminum to human nervous system. Studies have shown that aluminum has toxic effects on the nervous system. Excessive contact and accumulation of aluminum may lead to neurodegenerative diseases such as senile dementia and dialysis encephalopathy; the harm of bones, excessive intake of aluminum, will deposit in the bones, so that blood phosphorus reduces bone decalcification, resulting in bone softening, bone pain, muscle weakness, and other symptoms. Therefore, the analysis of soil aluminum pollution angle, the remaining sludge is not suitable for land use and greening mud.

Uranium and thorium are natural radioactive elements of the lanthanum series and are widely distributed in nature. If the contents of uranium and thorium in the excess sludge are higher than that of uranium and thorium in the soil, it will pose great harmless to animals and human beings. By analyzing the uranium and thorium elements in Cq sludge, the contents are 1.589 and 0.498 mg/kg, respectively, which were lower than the mean contents of uranium (3.03 mg/kg) and thorium (13.8 mg/kg) in Chinese soil and the mean contents of uranium (2.7 mg/kg) and thorium (9.4 mg/kg) in American soil [53].

Table 5. Element content of Cq sludge sample.

Element	Dried Cq Sludge (mg/kg)	Cq Sludge Ash (mg/kg)	Limit Value		
			Land Improvement [51]	Mixed Landfill Mud [50]	Greening Mud [52]
Be	0.877	N.D.	—	—	—
B	8.35	28.23	100	—	150
Al	6434.48	15,344.93	—	—	—
Sc	2.65	2.73	—	—	—
Ti	290.439	1065.99	—	—	—
V	34.274	122.62	—	—	—
Cr	24.71	96.24	600	1000	600
Mn	164.189	518.72	—	—	—
Fe	6806.09	24,049.88	—	—	—
Co	4.824	18.86	—	—	—
Ni	41.61	150.44	100	200	100
Cu	18.33	67.70	800	1500	800
Zn	628.958	2300.92	2000	4000	2000
Ge	1.345	4.08	—	—	—
As	86.723	301.03	75	75	75
Se	598.41	719.43	—	—	—
Mo	3.427	13.20	—	—	—
Sb	53.207	175.39	—	—	—
Ag	0.188	0.56	—	—	—
Cd	0.287	0.99	5	20	5
Hg	3.452	0.27	5	25	5
Pb	9.715	38.69	300	1000	300
Th *	0.50	1.77	—	—	—
U *	1.59	5.90	—	—	—

Note: *: radionuclides.

3.4. Analysis of Solid Waste Landfill

According to the risk analysis of excess sludge, the residual sludge from the refinery in this study is general solid waste. If it is landfilled, it needs to be landfilled to a general solid waste landfill. The excess sludge production is large, the water content of which is high, remaining 80–85% after mechanical dewatering, thus taking up large spaces. In addition, sludge with a high water content lacks the geotechnical stability necessary for landfill disposal, which causes certain difficulties in landfill operations [54,55]. Germany first specified the geotechnical properties of sludge landfill disposal [56]. The sludge should be filled with a shear strength of not less than 25 kPa, or an unconfined compressive strength of not less than 50 kPa. In China's "Technical Specification for Domestic Waste Sanitary Landfill Treatment" (GB 50869-2013) [57], the geotechnical indices for sludge landfill must fulfill the following conditions: the shear strength of the cross plate is not less than 25 kPa, and the unconfined compressive strength is not less than 50 kPa. Zhang Hua et al. [54] found that the sludge moisture content can be reduced to about 64% to meet the rockfill soil mechanics index. When the moisture content of Cq residual sludge drops to about 62%, it can meet the rockfill soil mechanics index. However, China does not give specific provisions on geotechnical indicators for industrial solid waste landfill disposal. In China's "General Industrial Solid Waste Storage and Disposal Site Pollution Control Standards" (GB18599-2001) [58], when solid waste with high humidity is required to be landfill, pretreatment, or adsorption desiccant is required.

The excess sludge has a high content of organic components and a high content of C, H, O and N. For example, the contents of C, H, O, and N in Cq sludge are shown in Figure 2. Landfill disposal is easy to cause anaerobic degradation of sludge. It produces gases, such as CH_4 and CO_2, which have certain negative effects on the environment.

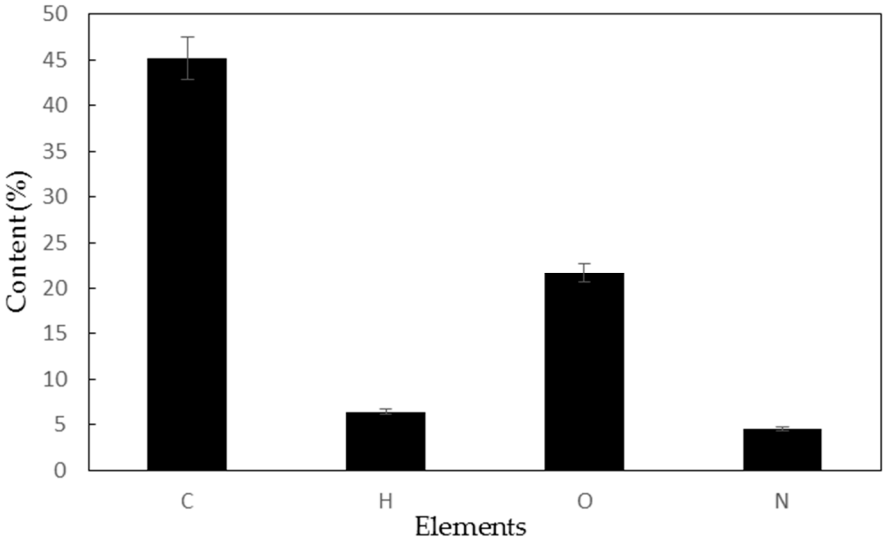

Figure 2. Contents of C, H, O, and N in Cq sludge samples.

3.5. Analysis of Petrochemical Excess Sludge Incineration

The sludge has different water content, and the amount of auxiliary fuel required for incineration disposal and the amount of incineration flue gas generated are quite different. When the dry sludge has a calorific value of 3500 kcal/kg and a water content of 33.3–80%, the sludge with a dry sludge content of one ton is incinerated. When the incineration temperature is maintained at about 800 °C, the required auxiliary fuel oil is between 0 and 850 kg, the self-sustained combustion of sludge can be achieved when the water content is less than 33.3%, and the amount of flue gas generated by incineration is about 7000–25,000 m^3 [59]. The higher the moisture content of the sludge, the more auxiliary fuel oil required for incineration, and the greater the amount of flue gas generated, hence the

heavier the tail gas treatment burden. Therefore, sludge incineration needs to be dried to reduce the moisture content of the sludge.

Drying methods for sludge include convection, conduction, and radiation, or a combination of these technologies. However, no matter which way, sludge has serious natural and dust explosion risks when it is hot-dried. When the solid content of the sludge is more than 80%, it is finely granulated and has a small particle size. The frictional collision between the sludge and the surrounding medium can generate a large amount of dust with a particle size of less than 150 μm. The dust has a certain oxygen content. Dust explosion is highly prone to occur under conditions such as temperature. Generally, the lower limit of the explosive concentration of organic dust is 20–60 g/m^3. When the protective gas is nitrogen, carbon dioxide, and water vapor, the minimum oxygen content of the dust combustion explosion is about 5%, 6%, and 10%, respectively. Therefore, the sludge is generally dried to a moisture content of about 30%.

The petrochemical excess sludge contains a certain amount of petroleum hydrocarbons. For example, the Cq wet sludge has a petroleum hydrocarbon content of 0.564%. When the sludge drying process is selected, the petroleum hydrocarbon content in the gas phase during the drying process needs to be analyzed to prevent flash-explosion.

The sludge contains elements such as N, C, S, and P. The contents of elements such as N, C, S, and P of Cq sludge are shown in Figures 2 and 3, respectively. Gases such as CO, CO_2, NO, NO_2, P_2O_3, P_2O_5, SO_2, and SO_3 are easily generated during incineration, and the corrosion dew point of the flue gas containing COx-NOx-SOx-POx-H_2O system is related to the thermodynamic properties of the mixed gas components. The composition fluctuates greatly, causing serious corrosion of the equipment.

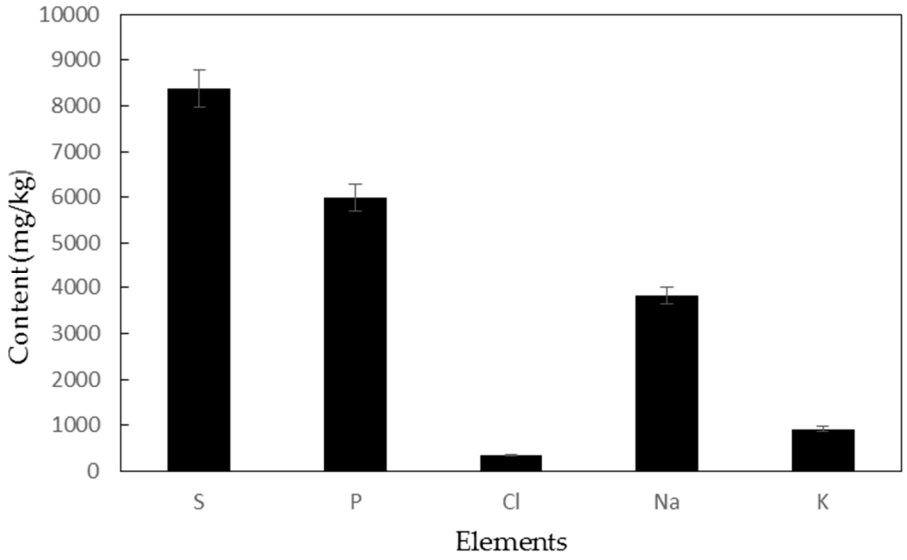

Figure 3. Contents of S, P, Cl, Na, and K in Cq sludge.

As can be seen from Figure 3, the content of Cl in Cq sludge is 353 mg/kg. The contents of Na and K are shown in Figure 3. The melting point of NaCl and KCl is low, and the sludge is easy to follow the flue gas during incineration. Entering the exhaust gas, it is easy to react with sulfur oxides in the presence of water and oxygen to form HCl and Cl_2, which is highly corrosive to the equipment.

To avoid the generation of dioxin, the temperature of incineration and disposal of solid waste is generally not lower than 850 °C [49]. In this study, heavy metal elements of ash obtained from the incineration of Cq sludge at 850 °C is shown in Table 5. It can be seen from Table 5 that the content of Ni, Zn, and As elements exceed the standard. In addition, the contents of uranium and thorium are 5.90 and 1.77 mg/kg, respectively, which are lower than the mean contents of uranium (3.03 mg/kg) and thorium (13.8 mg/kg) in Chinese soil [53]. Furthermore, from the change of Hg content before and

after sludge combustion, i.e., the content of Hg in Cq sludge (dry, ash content: 23.1%) is 3.45 mg/kg, while the content of Hg in sludge ash is 0.27 mg/kg, it was found that about 92% of Hg is eliminated by flue gas, that is, the emission of Hg from incineration sludge is 3.18 g/t. Note that the Hg emission in the Japanese sludge incineration disposal system is controlled to 0.2 g/t [60]. Hg emitted into the atmosphere can enter oceans and terrestrial waters directly from the atmosphere or even from its deposits in the surrounding basin without a particular source of Hg [60,61]. Therefore, the Hg in flue gas after sludge incineration must be disposed.

4. Conclusions

In this work, a novel classification and disposal strategy of excess petrochemical sludge was proposed combined with the physical and chemical properties of sludge and disposal analysis. The feasibility of the sludge classification and disposal strategy was verified by studying the three petrochemical residual sludge samples of Ah, Bl, and Cq. The results show that the indicators of Ah, Bl, and Cq wet sludge are lower than the limit value of hazardous waste. Then, taking Cq sludge as an example, the possibility of landfill, soil remediation and greening mud was analyzed. The elemental content was compared with the limit value of China's standards for landfill, soil remediation, and greening. Natural radioactive elements uranium and thorium in Cq sludge were analyzed, the contents of which are 1.589 and 0.498 mg/kg, respectively, lower than the mean contents of uranium (3.03 mg/kg) and thorium (13.8 mg/kg) in Chinese soil. From comparison, it was found that the Cq sludge is not suitable for landfill, soil remediation, and greening mud because of excessive arsenic. Finally, Cq was taken as an example to analyze the risk characteristics of petrochemical residual sludge landfill and incineration. The landfill risk of the petrochemical residual sludge solid waste landfill is mainly reflected in landfill gas, resulting in higher maintenance costs of the petrochemical excess sludge solid waste landfill. The risk of Cq sludge incineration involves dust explosion during the sludge drying process, petroleum hydrocarbon flash explosion, and equipment corrosion that is caused by the production of HCl, Cl_2, and COx-NOx-SOx-POx-H_2O due to high temperature reaction.

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