Experimental and feasibility study on co-incineration of sewage sludge and municipal solid waste

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Abstract

At present, the co-incineration technology of sewage sludge and municipal solid waste (MSW) has developed rapidly due to its advantages of reduction, harmlessness, and resource utilization, but the heating value, organic content, and heavy metal content of the sewage sludge will affect the co-incineration process. Therefore, the composition of sewage sludge needs to be clarified first. This study investigates some national surveys that provide a representative elemental composition of sewage sludge and analyzed 119 sewage sludge samples from 32 wastewater treatment plants in Japan. The different seasons, sewage collection systems, dewatering methods, coagulants that are added or not, digested or undigested of sewage sludge were investigated. The element content in sewage sludge varies widely and has large deviations, but concentrations of constituent elements in sewage sludges from different countries were similar. However, the sewage collection system, digestion process, and the coagulant used for dewatering affect the performance and element concentration of the dewatered sludge. Seasonal effects and dewatering devices have little effect on the concentration of elements. The digestion process has the strongest correlation with the elemental composition of dewatered sludge, especially organic matter. According to the sewage collection system and digestion status, the standard components of these four types of sludge are proposed. The heavy metal content of the sludge from the combined system is higher than that of the sludge of the separate system, and the heavy metal content of the digested sludge is higher than that of the undigested sludge. Undigested sludge has greater agricultural recycling potential than digested sludge, but the Cu concentration of dewatered sludge in Japan exceeds the regulatory limit for agricultural use, and caution is required. According to Lafarge Cement Polska's standards, the sewage sludge in Japan can be used as alternative fuels in the cement industry. Moreover, the composition of sewage sludge is similar to RDF, and almost meets the Japanese Industrial Standard Biomass Solid Fuel-15 (BSF-15, a heating value higher than 15 MJ/kg).

From the above research, it was found that sewage sludge has a higher heating value and has considerable advantages as a biomass fuel. However, heavy metals are a factor that requires careful attention. Therefore, this study explored the effects of co-incineration of sewage sludge and MSW on heavy metals including Cu, Pb, and Cd. The results show that the incineration of MSW is mainly caused by organic matter and fixed carbon but mainly caused by volatiles in sewage sludge, and the interaction effect in the co-incineration process which delays the pyrolysis and incineration of polymer and fixed carbon. The residual ratio of heavy metals in the mono-incineration of sewage sludge was higher than that of MSW, which is inferred that the Cl content in MSW is much higher than that in sewage sludge, so it is possible that heavy metals and Cl form metal chlorides and volatilize. The interaction effect of co-incineration can be seen in Cu, but not in Pb and Cd. XAFS analysis proves that Cu in the ash content of MSW exists in the form of $CuO(s)$, but in the form of $Cu_3(PO_4)_2$ in the ash of sewage sludge, $Cu_3(PO_4)_2$ was also found in co-incineration ash. $CuO(s)$ is relatively unstable and may form $CuO(g)$ or $CuCl(s)$ and volatilize with high temperature and Cl presence. It can be seen that P in the sludge has a stable effect on Cu during the co-incineration process.

In addition, from the perspective of development potential, co-incineration is an advantageous method for countries with a small land area. There have been many examples of co-incineration of sewage sludge and MSW in Japan. Taiwan is currently in the trial stage of sewage sludge and MSW co-incineration. This study conducted a feasibility assessment of co-incineration in Taiwan. According to the questionnaire, literature, and government policies, set the necessary conditions for cooperative incineration, including operation ratio < 90%, mixing ratio < 3% for dewatered sludge and 1% for dry sludge, and the actual distance within 30km between the wastewater treatment plant (WWTP) and municipal solid waste incinerator (MSWI). According to the drying equipment of the WWTP, 4 scenarios were set up and selected in sequence according to the necessary conditions. 39 feasible combinations were screened out, accounting for 89.7% of the national sludge production which can be co-incineration. Since the amount of sewage sludge is very small compared to MSW, the operating ratio and mixing ratio hardly affect the potential of co-incineration. Only the distance between the WWTP and MSWI is a key factor. Compared with the co-incineration of dried sludge, dewatered sludge had fewer greenhouse gas emissions. Although the co-incineration of dried sludge can reduce greenhouse gas emissions during transportation and incineration, the power generation during the incineration process can offset greenhouse gas emissions. However, the drying process produced a large amount of greenhouse gas emissions, and overall, still causes a large amount of greenhouse gas emissions.

Contents

Chapter 1 Introduction and literature review

1.1 Background

Sewage sewer construction is regarded as one of the indicators of national or urban progress [1], indicating that it is an indispensable infrastructure. With the progress and development of cities, sewage treatment is increasing, and bringing a large amount of sewage sludge production. Sewage sludge is the residue generated in the sewage treatment process [2], which was landfilled in various countries, but its characteristics of containing a large amount of organic matter and nutrients have gradually been regarded as a potential biomass source for agricultural use.

Many studies have indicated that the fertilization of sewage sludge has a positive effect on the improvement of soil quality [3][4][5]. In addition to recycling as agricultural fertilizers, the recovery of these elements from sewage sludge has also become a topic of concern [6][7][8][9]. However, the composition of sewage sludge is easily affected by sewage sources and the treatment process of the wastewater treatment plant (WWTP). The content of heavy metals in the sewage may be concentrated and accumulated in the compost after the bioconversion process. There have been several cases where high heavy metal concentrations in crops or groundwater have caused serious disease due to accumulation in the human body [10][11].

Therefore, many countries have restricted the land use of sewage sludge. For example, there are strict standards for agricultural use of sewage sludge in the UK and Germany [12]; Switzerland and Norway also prohibit the agricultural use of sewage sludge [13]. Although the heavy metals in sewage sludge may cause harm during land use, extracting and recovering heavy metals from it also has an economic value from another perspective [14]. In addition, the organic matter in sewage sludge can be made into biomass fuel through processes such as thermal drying or carbonization. The previous study has shown that the concentrations of organic elements like carbon (C), hydrogen (H), and oxygen (O) have a strong relationship with the heating value, which is the most important parameter for the incineration of fuels [15].

Sludge incineration is to oxidize and decompose the organic matter in sludge into carbon dioxide and water under high-temperature conditions. At the same time, the waste heat generated during the incineration process can also be recovered to realize the rapid reduction, harmlessness, and resource utilization of sludge [16]. Sludge incineration has been in development, the first recorded sludge incinerator was installed in 1930 in a midwestern sewage plant in the United State [17]. Sludge incineration has become a choice of sludge treatment method in many developed countries and newly industrialized countries [18][19]. For countries or regions with narrow land and dense population, incineration is a faster and economical method compared with composting and agricultural utilization. Especially in Japan, sludge incineration technology has become a common treatment [20]. In addition, the incineration of sludge as biomass energy has the considerable potential [21].

Sludge incineration is a means to achieve the maximum reduction of sludge. Due to sludge

being rich in organic matter, its heating value can also provide the energy required for incineration. According to the heating value of the sludge, it can conduct mono-incineration or co-incineration [22]. Mono-incineration of sludge requires technology and equipment, as well as high capital cost and environmental impact assessment. The use of existing incinerators such as coal-fired power plants, cement plants, municipal solid waste incinerators (MSWI), etc. for co-incineration can greatly reduce operation and maintenance costs. In addition, in most cases, sewage sludge only accounts for a small part of the capacity of the incineration plant, so that the process or product is not adversely affected [23]. However, the existing incineration facilities were originally designed for specific substances such as coal, municipal solid waste (MSW), etc. The co-incineration of two different substances, different moisture content, and composition may cause interaction [24], especially the migration of heavy metals. Heavy metal compounds based on copper (Cu), lead (Pb), and zinc (Zn) are volatile at medium temperatures and have sufficiently boiling points that they can become gaseous and diffuse into the atmosphere during incineration, causing air pollution [25][26]. Whether this interaction has an impact on the equipment and the environment must be considered. Moreover, whether the operating condition of existing facilities has sufficient capacity to receive sludge also needs to be considered.

1.2 Co-incineration of sewage sludge

Co-incineration refers to the incineration of two or more substances in the same incinerator. Many studies have pointed out that co-incineration changes the incineration behavior of the boiler, affects the composition of substances during incineration. Moisture, volatiles content, ash content, constituent elements, and mixing ratio also bring some effects to co-incineration [27]. For example, high moisture content causes unstable incineration [28]; heavy metals and chlorine content increase the volatilization of heavy metals [29]. Co-incineration may also increase the emission of harmful gases, the presence of nitrogen (N), sulfur (S), chlorine (Cl) provides the precursors required for the formation of nitric oxide, nitrous dioxide, sulfur dioxide, hydrogen chloride, or dioxins [30][31][32]. The migration of heavy metals will be affected by temperature, moisture, and some elements such as phosphorus (P), Cl, S, which cause volatilization or precipitate [33][34]. The alkaline earth metals may cause slagging, corrosion and deterioration during the incineration process [35]. The composition of different substances leads to a interaction effect in the co-incineration process, and the interaction effect promotes the volatilization in the smoker or residual in slag also affects the treatment methods. Therefore, the characteristics and proportions of the co-incineration substances play a decisive role in the result. Table 1.1 sorts out the comparison of three common substances used in co-incineration, including coal, biomass (sewage sludge, agricultural waste), etc. [36].

Table 1.1 Comparison of common substances used in co-incineration [36].

Element	Coal	Biofuel	Waste
S	Medium to high	Low	Low to medium
Cl	Medium	Low to high	Medium to high
K	Bound	Medium to high	Low
Other alkalies	Normal	Low	Low to normal
Al, Si*	High	Low to high	High

*Al: aluminum, Si: silicon.

Compared with other materials, coal has a high S content, some coals, biomass, and wastes have high Cl content, and the role of potassium (K) in biomass in the formation of sediments during incineration. Therefore, the interaction effect caused by the different compositions of the co-incineration objects needs to be clarified. When evaluating the suitability of co-incineration, the composition of the material is very important.

The composition of sludge is different in various countries, even in different areas within the same country [37], which is varies depending on the source of sewage, region, season, and the treatment process of the sewage plant [38]. Thus, several studies have investigated sewage sludge composition at the national level. Eriksson et al. [39] at the Swedish Environmental Protection Agency investigated 61 trace elements in sewage sludge from about 50 WWTPs, provided average elemental concentrations, and studied the effects of these elements on agricultural use. Vriens et al. [40] assessed the concentrations of 69 elements in digested sewage sludge and effluent samples from 64WWTPs as well as in major rivers in Switzerland. The United States Environmental Protection Agency collected 66 sewage sludge samples from 74 randomly selected WWTPs in 36 states to determine the main organic elements (C, H, N, O, and S) of the biosolids in the Targeted National Sewage Sludge Survey [41]. Moreover, the United States National Sewage Sludge Repository collected sewage sludge from 74 WWTPs and analyzed 28 metals [42]. These surveys provide an important source of reference.

As for the application of co-incineration, some European countries and the United States have gradually co-incineration solid biomass fuels with coal to reduce the impact on air pollution and move towards conversion to a 100% biomass-fired system. However, for the treatment of sewage sludge, the United States has put forward the priority order of land use, incineration, and landfill in the 40 CFR Part503, and has strict specific requirements for incineration. In Germany, among the biomass co-incineration power plants, 50% of the plants use sewage sludge as fuel, and the co-incineration ratio can reach 3% without significant impact on the incinerator [43]. According to the literature, the incineration equipment and devices of the coal-fired power plant and MSWI are different, so the situation of co-incineration will also be different [44]. Sewage sludge and MSW are wastes produced in human life and generated in cities, most MSWIs are equipped with power generation equipment to recovery the incinerated waste heat for reuse. So, the cooperation between the WWTP and MSWI is worth looking forward to. And the complete flue gas treatment system of the MSWI can properly treat the gas

generated during the co-incineration [45]. In addition, the research [46] reported that the components in the sludge incineration are easy to form stable substances, so the interaction effect of co-incineration may stabilize the substances in the incineration residue and without being released into the environment, making the co-incineration of sewage sludge and MSW become a development direction [47].

1.3 Sewage sludge and MSW management in various countries

1.3.1 United States

In the United States, to solve the problem of sewage sludge, Standards for the Use or Disposal of Sewage Sludge (40 CFR Part 503) was promulgated in 1991, which regulates the requirements for the utilization or disposal of sewage sludge, including general requirements, pollutant discharge limit values, management and operation regulations [48]. The disposal of sewage sludge in the United States is mainly based on land application, incineration, and landfill. According to the providing data from 2004 [49], there were 16,583 WWTPs, which were estimated to produce 6.5 million tons of sewage sludge (dry basis) in the United States. And based on research published in 2017 [50], which estimated that the production of sewage sludge is 13.84 million tons/year.

Figure 1.1 shows sewage sludge disposal in the U.S [50]. In 2004, 41% were applied to farmlands, 12% were treated to exceptional quality standards and publicly distributed for various uses, 2% were used in land reclamation, silviculture, or other uses, 28% were landfilled, 2% were put in surface disposal units, and 15% were incinerated. And in 2019, 51% were land application, 10% were other management practices, 22% were landfill, 1% were surface disposal, and 16% were incineration. It can be seen that over the past 15 years, the proportion of sewage sludge landfills has decreased, and land application has increased.

Figure 1.2 shows the MSW treatment in the U.S. from 1960 to 2018 [51]. The amount of MSW in the U.S. was increasing year by year. In 2018, the amount of waste generated was about 265 million tons, and of the MSW generated, about 23.6% were recycled and 8.5% were composted, which was equivalent to a 32.1% recovery ratio. In addition, nearly 11.8% were combusted with energy recovery and 50% of MSW were landfilled. The first MSW incinerator in the U.S. was built in 1885, and the incineration of MSW increased in the 1980s. By the early 1990s, the U.S. had incinerated more than 15% of all MSW generation [52]. In 2019, 67 power plants incinerated about 25 million tons of MSW and generated about 13 billion kWh of electricity [53].

2004

Incineration 15%

Surface disposal unit 2%

Landfill 28%

Land reclamation or silviculture 2%

Farmland 41%

Exceptional qulality standards 12%

2019

Incineration 16%

Surface disposal 1%

Landfill 22%

Land Application 51%

Other management practices 10%
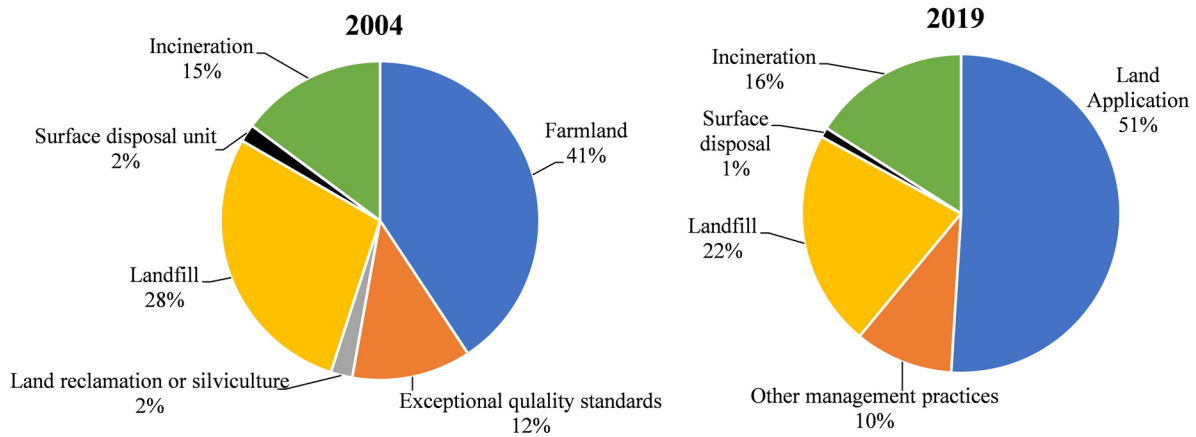
Figure 1.1 Sewage sludge disposal in the U.S [50].
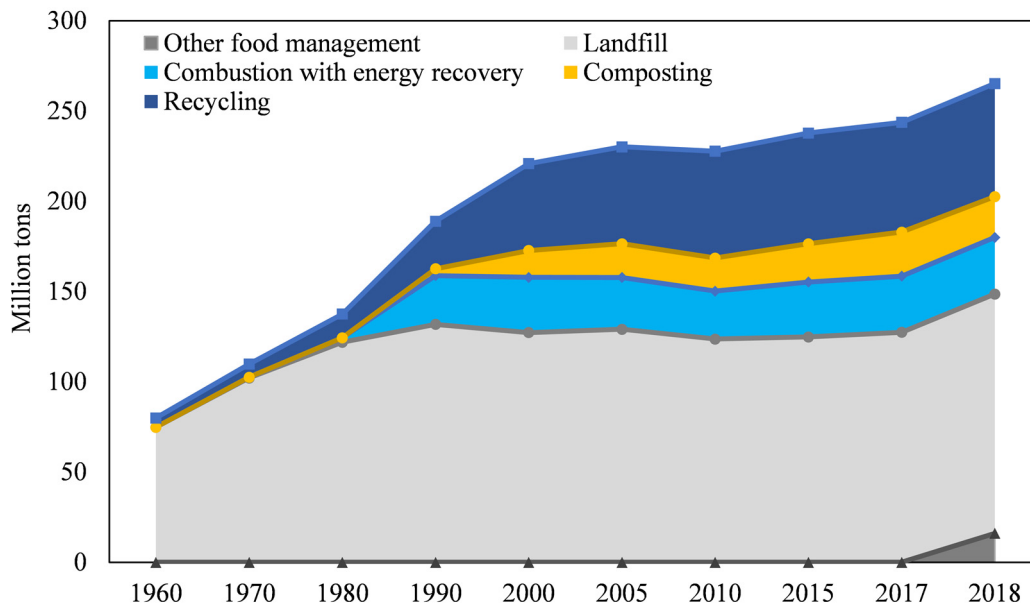
Figure 1.2 MSW treatment in the U.S. from 1960 to 2018 [51].

1.3.2 EU

According to European statistics, the amount of sewage sludge produced in Europe was about 10 million tons (dry basis) in 2016, however, because European countries have different sewage treatment systems and management policies, and the statistics data of some countries is not completely collected, the actual sewage sludge production may be more than the statistical data (the sludge production of Belgium, Denmark, Italy, Finland, Norway, Switzerland, and the UK were not available in 2016) [54].

Figure 1.3 shows the sewage sludge disposal in European countries which according to the latest statistics of each country [54]. Incineration and agricultural use are the most widely used sewage sludge disposal in the EU, however, the disposal methods used by countries vary greatly. More than half of the sewage sludge in five countries including Switzerland, Netherlands, Belgium, Germany, and Austria were treated by incineration because the agricultural use of

sewage sludge is strictly restrained by regulations [55]. Among them, 100% of the sewage sludge in Switzerland was treated by incineration; It was mostly used for agriculture use in Demark, UK, Spain, Norway, Bulgaria, and Ireland; In Malta, Serbia, Bosnia, and Herzegovina, all sewage sludge were landfilled, in addition, Croatia, Turkey, Italy, Romania also mainly treated sludge by landfill.

MSW generation totals vary considerably, ranging from 280 kg/capita in Romania to 844 kg/capita in Denmark [56]. The variations reflect the difference in consumption patterns and economic wealth, but also depend on the collection and management of the MSW. Figure 1.4 shows the MSW treatment in the EU-27 from 1995 to 2019 [56]. On the whole, although the EU is generating more MSW, the proportion of landfills was decreasing from 61% in 1995 to 23% in 2019. The incineration of MSW had also grown from 15% to 27%. The Directive 31/1999 on landfilling had stipulated that member states should reduce the organic fraction of MSW sent to landfills, resulting in the recovery ratio has grown from 19% to 48%. Figure 1.5 shows the distribution of 450 MSWI [57] and the incineration capacity of the entire country in the EU countries in 2011. The difference in MSW incineration of the member states of the EU could be seen from the Figure. Denmark has the highest incineration amount, followed by the Netherlands, Norway, Sweden, and Switzerland, and the remaining countries are in the middle level, and some countries do not have MSWI.
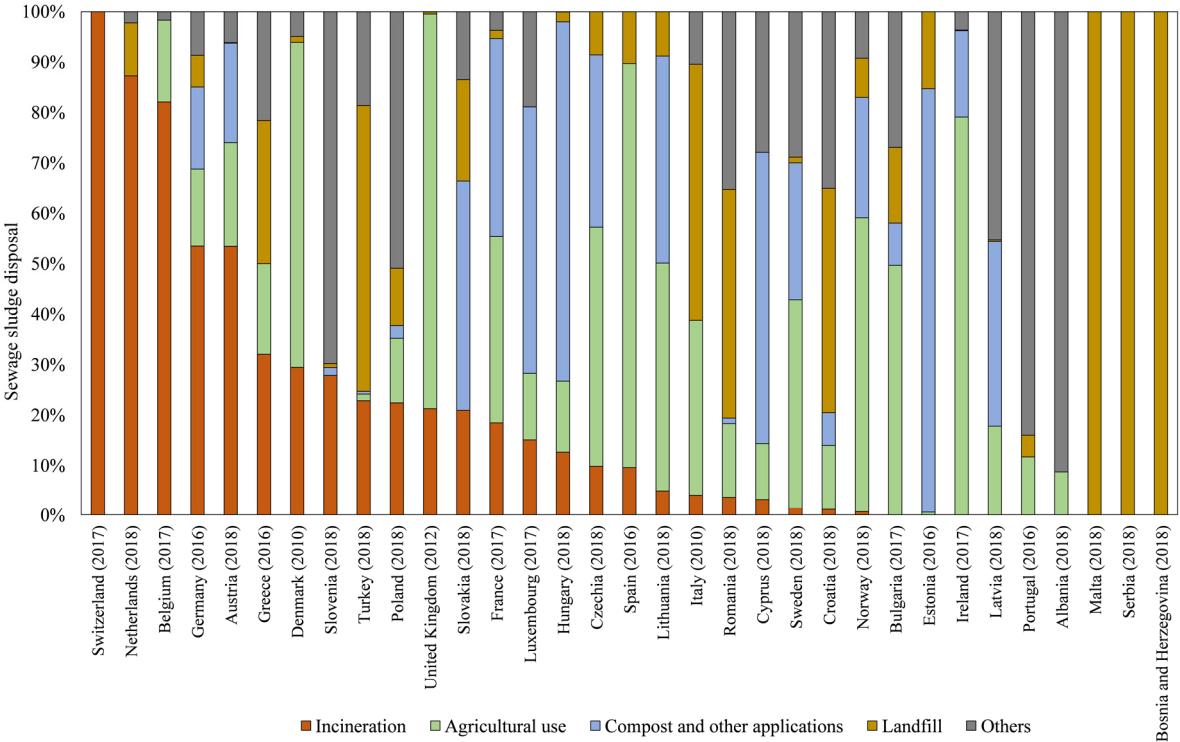
Figure 1.3 Sewage sludge disposal in European counties [54].
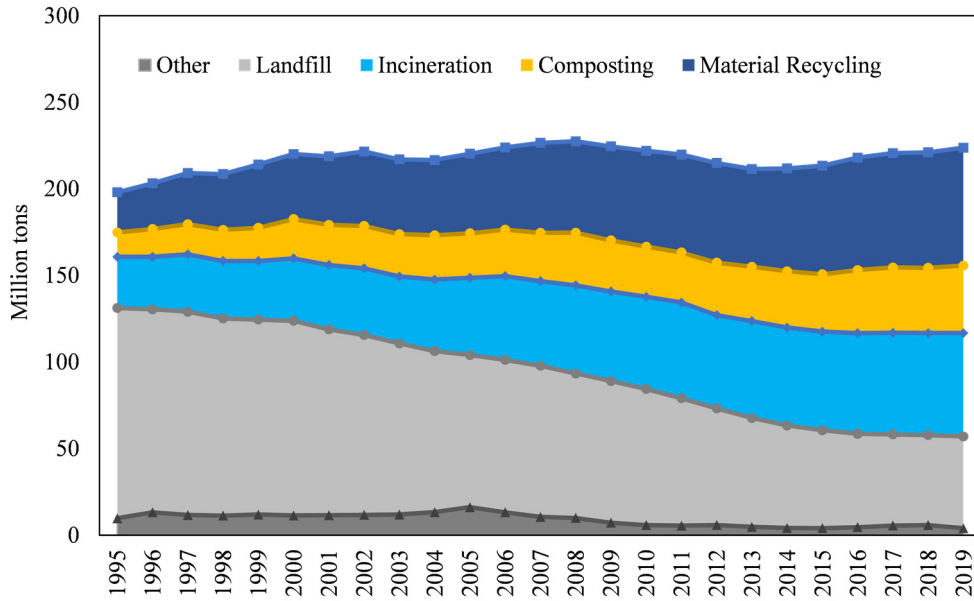
Figure 1.4 MSW treatment in the EU-27 from 1995 to 2019 [56].
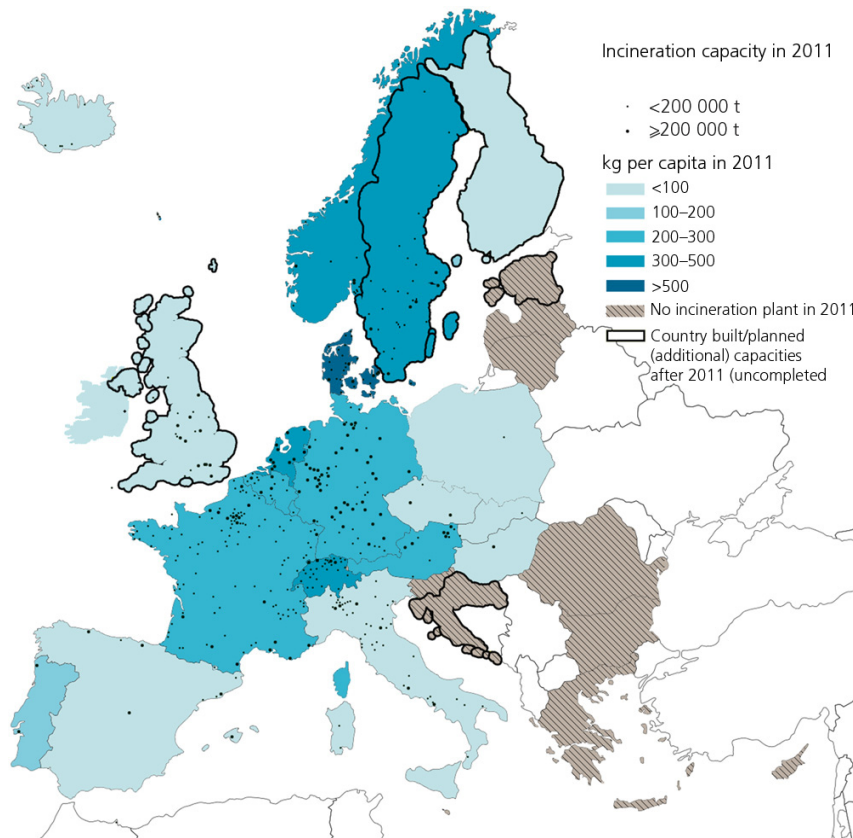
Figure 1.5 Distribution of 450 MSWI in the EU countries [57].

1.3.3 China

In China, by the end of 2019, there were 5,476 WWTPs in operation, with an annual production of 7.81 million tons of sludge (dry basis). China has a large land area, and the sewage sludge treatment and disposal technologies used in different regions vary greatly. For example, composting is the main method applied in South China, while incineration is the main method applied in East China. In contrast, landfill is mainly used in other areas.

Figure 1.6 shows the contribution of different sewage sludge disposal approaches in China from 2009 to 2019 [58]. In the past, land application was the mainstay technical route of sludge disposal in China, but in recent years, the contribution of sewage sludge disposal had a dramatic change. In 2019, about 29.3% of the sludge was disposed of by land application, 26.7% by incineration, 20.1% by sanitary landfills, 15.9% by building materials utilization, and 8.0% by other disposal methods. In 2016, China produced more than 10% of the MSW generation in the world, and it has continued to increase. Figure 1.7 shows the generation and treatment of MSW in China. The MSW generation has maintained an upward trend, it had risen to 228 million tons in 2018. The landfill had been the main treatment for MSW in China in the past few decades. By 2018, the number of landfill sites has exceeded 660 plants [59]. However, with the development of waste incineration technology and the utilization of its resources, waste incineration has begun to receive attention. In 2004, there were only 54 MSWIs in China, with a designed annual processing capacity of 6.17 million tons, processing 2.9% of the collected MSW (4.49 million tons). By 2018, there were 331 MSWI with a designed annual capacity of 133 million tons, processing 44.67% of the collected MSW (102 million tons), and at least 80 of MSWI (waste-to-energy, WTE) are under construction [60]. Even so, most of the MSWIs are located in eastern China, and the development of incineration is quite uneven as shown in Figure 1.8 [61].
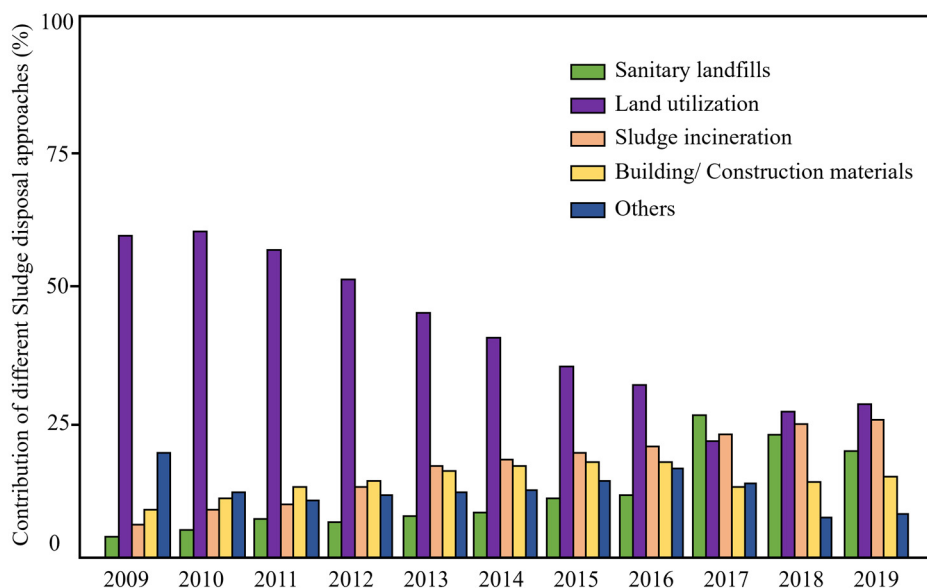
Figure 1.6 Contribution of different sludge disposal approaches in China [58].
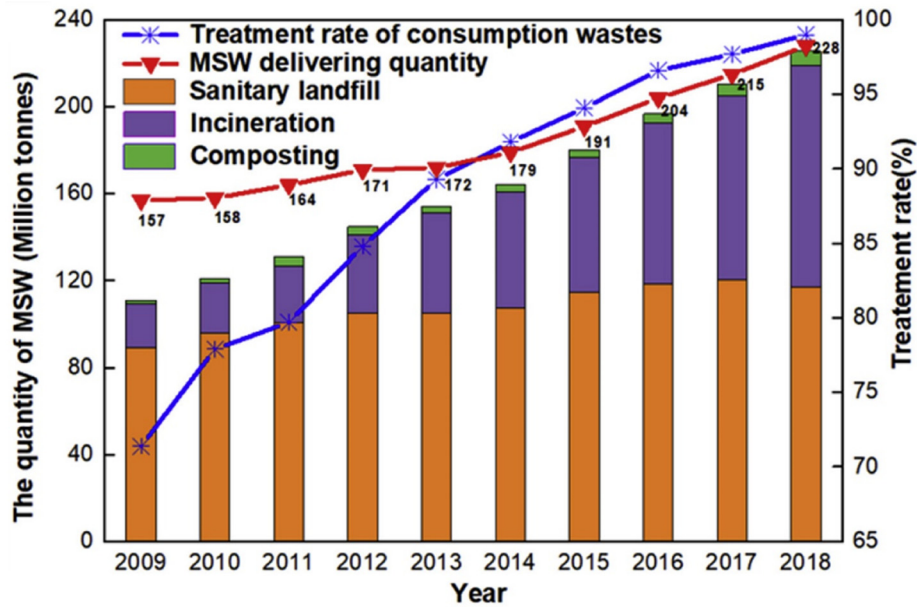
Figure 1.7 MSW generation and treatment in China from 2009 to 2018 [59].
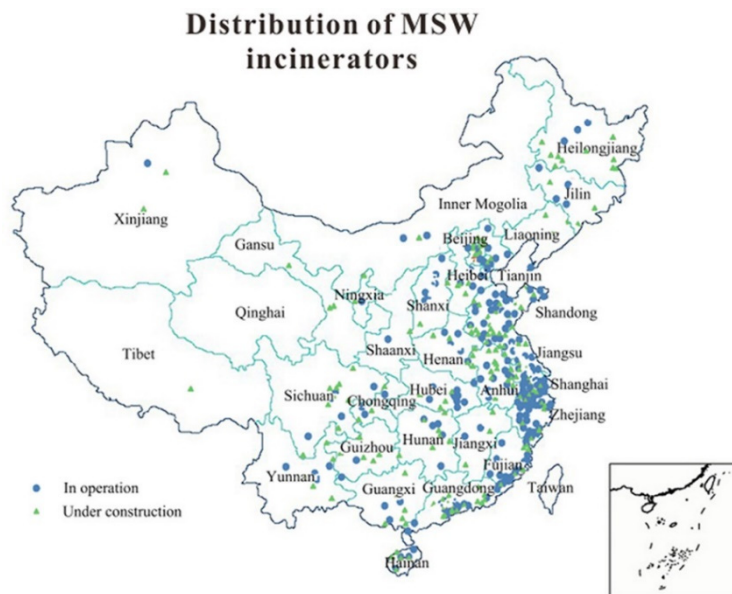
Figure 1.8 Distribution of MSWI in China in 2015 [61].

1.3.4 Japan

Figure 1.9 shows the generation and treatment of sewage sludge in Japan [23]. There are currently about 2,200 sewage treatment plants in Japan. The sewage sludge generation and recycling rate have grown steadily in the past in Japan. In 2010, the sewage sludge generation was 2,268 tons (dry basis) and the recycling rate was as high as 78%. However, when the Fukushima nuclear disaster occurred in 2011, the recovery rate dropped sharply to 55%. After that, the amount of sewage sludge produced stabilized and the recovery rate slowly recovered. In 2019, the sewage sludge generation was 2,341 tons (dry basis), the recovery rate has risen to 75%. The landfill was the main method in the past, but the recycling rate exceeded 50% after

1999. The recycling includes construction material (21%), cement (31%), green farm (14%), fuel (8%), and others (1%) in 2019, only 23% of the sewage sludge remains in landfills. In 2019, there were 1,067 MSWIs in Japan (and 34 were under construction), with a capacity of approximately 176,707 tons/day. Figure 1.10 shows the MSW generation and treatment in Japan from 2010 to 2019. Since 2012, the total MSW generation had decreased slightly but remained flat for the past few years. The MSW generation in 2019 was 42.74 million tons. Among the intermediate treatment, the incineration was 32.95 million tons and accounted for 80.5%.
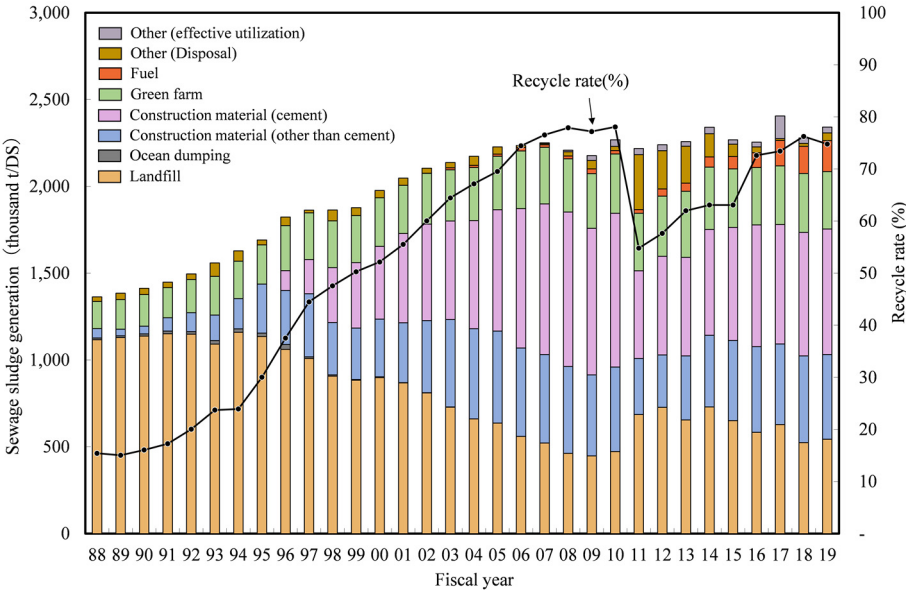
Figure 1.9 Generation and treatment of sewage sludge in Japan [15].
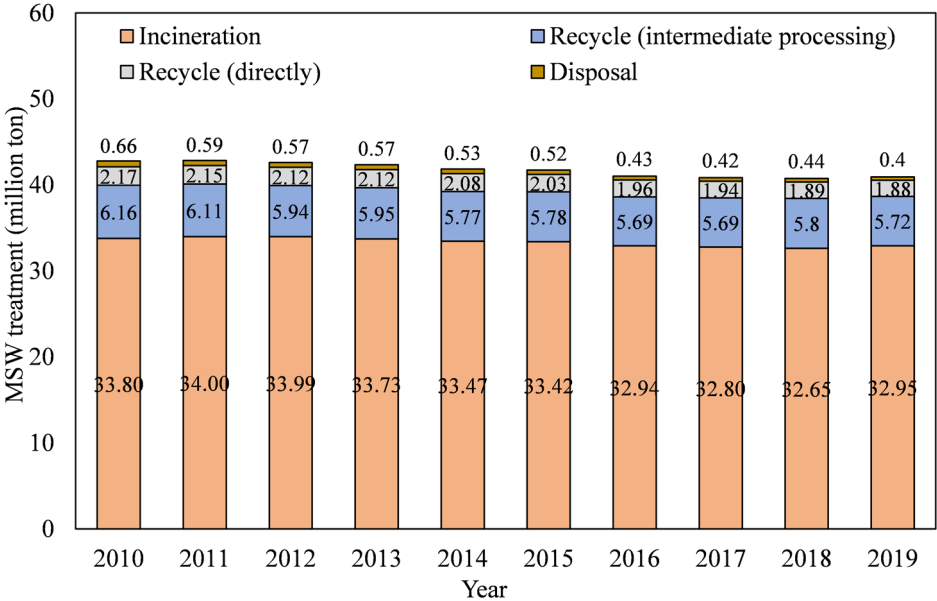
Figure 1.10 MSW generation and treatment in Japan from 2010 to 2019 [15].

1.4 Research on co-incineration of sewage sludge and MSW

Many studies have conducted experiments on sewage sludge and MSW co-incineration of laboratory-scale research. In South Korea, Lee et. al [62] reported that the co-incineration reaction increases with the decrease of the water content of the sludge in the case of co-incineration. With the increase of sludge particle diameter and water content, the decomposition rate decreased, and the activation energy increased. These properties are more affected by particle size than by moisture content.

Lee et. al [63] also studied the feasibility of refuse-derived fuel (RDF) as an alternative fuel and co-incineration with sewage sludge. The results showed that as the incineration temperature and time increase, the carbon oxide concentration decreases, but the carbon dioxide, nitrogen oxide, and sulfur oxide concentrations steadily increase. The efficiency of co-incineration with sewage sludge and RDF is much higher than that of mono-incineration of sewage sludge. In China, Chen et. al [45] studied the volatilization characteristics of heavy metals in the co-incineration of sewage sludge and MSW, which indicated that the volatilization efficiency of heavy metals is a linear function of temperature and a quadratic function of Cl content. The distribution of mercury (Hg) and the transformation of chemical forms during the co-incineration of sewage sludge and MSW have also been studied [64]. This study showed that when sewage sludge and MSW are co-incineration, Hg tends to be more concentrated in the ashes than that in mono-incineration. Co-incineration may also affect the chemical form of Hg in the ashes, thereby affecting the environmental risks of the ashes.

In addition, there are also some reports which studied the co-incineration on actual incineration plants. Biganzoli et. al [65] mentioned that the co-incineration of sludge and MSW is a very common practice in Europe. The study introduced a case of co-incineration of sewage sludge and chemical sludge in a WTE waste incineration power plant equipped with grate furnaces in northern Italy and compared the mass balance of toxic trace elements in co-incineration and mono-incineration. The results showed that the co-incineration of sewage sludge and chemical sludge did not lead to an increase in the total release of toxic trace elements in the environment, except for arsenic (As), which increases the bottom ash concentration by 5 times during the co-incineration. On one hand, As content had no change observed in the exhaust gas. This fact further ensures that the co-incineration of sewage sludge in the most advanced waste incineration power plants will not have a negative impact on the atmospheric environment. Toledo et, al [66] studied the behavior of six target heavy metals which including cadmium (Cd), Pb, chromium (Cr), Cu, Zn, and nickel (Ni) in the co-incineration of sewage sludge and MSW in a bubbling fluidized bed (BFB) in Spain. It showed that the amount or percentage of Cl in the raw material had an important influence on the fate of specific heavy metals such as Cd and Pb. And the fate of heavy metals depends on the residence time of ashes and particles at high temperatures, and these residence times vary depending on the type of incinerator may

be very different. In China, Chin et. al [67] carried out the co-incineration of sewage sludge and MSW in a medium-scale coal-filled incinerator, to study the effect of increasing the proportion of sewage sludge blending on co-incineration temperature and gas emissions, and also to evaluate the effectiveness of co-incineration in oxygen-enriched air. Moreover, which indicated that 30 wt.% of sewage sludge blended with MSW co-incineration is possible, and which with a moderate oxygen enrichment of 22 vol.% (dry basis) in the supply air.

Comprehensive assessment of co-incineration feasibility has also been studied. In Italy, Bianchini et. al [68] studied the assessment of waste heat generated from MSWI as the source used on sludge drying. It showed that thermal drying of sewage sludge is unsustainable if it is powered by primary energy, but it may be attractive if waste heat recovered from other processes is used. By recycling waste heat from MSWI as energy for drying sewage sludge, proper integration between WWTP and MSWI can be achieved.

Wang et. al [69] assessed the environmental impact of 4 sewage sludge treatment options in Taiwan which include carbonization, mono-incineration, direct landfill, and co-incineration with MSW. It indicated that co-incineration emits fewer greenhouse gases (GHGs) because the overall energy recovery rate of the electricity in the incineration process was higher than that in the carbonization process. Although this analysis considers heat recovery during coal substitution in the process of carbonization, power generation, and co-incineration, the energy used to dry dewatered sludge emits more GHG and contributes greatly to the types of damage caused by climate change. Zhang et. al [70] studied the feasibility of cooperation between WWTP and MSWI in Japan. It showed that about 39.3% of the sewage sludge in Japan has the potential to be sent to MSWI for co-incineration, and it can reduce GHG emissions and operating costs.

1.5 Summary

According to the above-mentioned background and literature, various signs indicate that sewage sludge generation will continue to increase. In the future, whether in developed or developing countries, many countries in the world must find suitable solutions to deal with the expected large amount of sewage sludge. The sewage sludge and MSW disposal methods of various countries used to be landfilled in the past. With the development of technology, treatment has also begun to change. In terms of sewage sludge, the treatment methods of sewage sludge in various countries have become quite diverse. The WTE incineration of sewage sludge has also become the director of development. In terms of MSW, although the amount of MSW generated in various countries is slowly increasing, the amount that needs to be disposed of is reduced by recycling and reuse. At present, incineration technology has been developed maturely, and incineration has become the mainstream way of disposing of MSW. Although some national surveys mentioned above provide the representative elemental composition of sewage sludge, the characteristic of dewatered sludge may be affected by seasons, sewage

collection systems, coagulants used for dewatering, and digestion processes. There has not had a national survey of sludge composition in Japan, and the factors affecting the composition of sewage sludge have not been clearly sorted out. In addition, although many researchers have investigated the effects of sewage sludge and MSW co-incineration, due to differences in the natural environment, geography, and living habits, the composition of sewage sludge and MSW are varying. Thus, the effect of co-incineration in Japan also needs to be compared and collated with the literature. Research in Taiwan has confirmed that the co-incineration of sewage sludge and MSW is efficient and harmless to the atmosphere [69]. In addition, Taiwan and Japan are located close to each other, and both belong to island countries in Asia. Co-incineration has proven to have considerable potential for sewage sludge treatment in Japan [70], and it is an advantageous treatment method for countries with a small land. Therefore, Taiwan should also have considerable potential for co-incineration as well as Japan.

1.6 Research objective and structure of the thesis

Based on the above background and literature, since national sewage sludge composition has not yet been investigated in Japan, the first purpose of this study is to investigate national sewage sludge composition, summarize the factors affecting the sludge composition, and propose a standard composition of the sewage sludge in Japan. Next, one of the investigated sludges to conduct a co-incineration experiment with MSW, and to understand the interaction effect on heavy metals by comparing mono-incineration and co-incineration. Finally, since the development and promotion of sewage sludge drying and co-incineration with MSW in Taiwan are conducted, the feasibility of national sewage sludge and MSW co-incineration and the effect of GHG reduction also be clarified in this study. The doctoral thesis consists of five chapters. The structure shows in Figure 1.11.

Chapter 1 introduces the development of sewage sludge co-incineration, the generation, and management of sewage sludge and MSW in various countries were summarized, as well as some previous research on the co-incineration of sewage sludge and MSW.

Chapter 2 investigates the elemental composition of 119 dewatered sewage sludge samples from 32 WWTPs in Japan, and the influential factors determining sewage sludge elemental composition were clarified. Through Hayashi's quantification method I, the relationship and influence of each element to the dominant categories were confirmed, and the correlation between the elements was also analyzed. The Van Krevelen diagram was used to determine the similarity in organic contents between sludge and biomass/fossil fuels. Principal component and cluster analyses were used to verify the identification of influential factors. Finally, a standard composition of dewatered sewage sludge in Japan was defined based on different sewage collection systems and digestion processes which shown to have the greatest influence on sludge composition.

Chapter 3 presents the sewage sludge and MSW co-incineration effect on the behavior of heavy

metals. Through co-incineration in different blending ratios (MSW, 10%, 20%, 50%, sludge), the effect of the co-incineration on the residual ratio of heavy metals in the ash was explored. Through TG-DTA analysis, the incineration behavior of sewage sludge and MSW and the effects of co-incineration were compared. The residual ratio of heavy metals including Cu, Pb, Cd, and P in the incineration ash was analyzed to understand the impact of the co-incineration on heavy metals. XAFS was used to indicate that the chemical status of Cu in incineration ash, and the influence of the co-incineration process on Cu could be clarified.

Chapter 4 elucidates the potential of sewage sludge and MSW co-incineration in Taiwan. A database of WWTPs and MSWIs in Taiwan was established. Through literature investigation to understand the current situation of co-incineration in Taiwan. 4 necessary conditions and 4 scenarios (according to the sludge is dewatered or dried), the potential of co-incineration nationwide in Taiwan has been evaluated. The GHG emission of all scenarios was compared. Chapter 5 summarizes the conclusion from these studies and proposes future works.

Chapter 1 Introduction and literature review
- Co-incineration of sewage sludge
- Sewage sludge and MSW management in various country
- Research on co-incineration of sewage sludge and MSW

Chapter 2 Survey of elemental composition in dewatered sludge in Japan
- Elemental composition of sewage sludge in Japan
- Comparison of element composition with other countries
- Factors affecting element content and distribution
- Evaluation of sewage sludge recycle

Chapter 3 Co-incineration effect of sewage sludge and municipal solid waste on the behavior of heavy metals
- Composition and characteristic analysis
- Behavior of mono-incineration and co-incineration
- Migration characteristics of heavy metal during co-incineration
- XAFS analysis of Cu chemical status
- Comparison of research in other countries

Chapter 4 Feasibility study on the co-incineration of sewage sludge and municipal solid waste in Taiwan
- Database of WWTPs and MSWIs in Taiwan
- Current situation of co-incineration in Taiwan
- Necessary conditions setting of co-incineration
- Potential of co-incineration
- Calculation of GHG emission

Chapter 5 Conclusion and future perspective

Figure 1.11 The structure of this doctoral thesis.

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Chapter 2 Survey of elemental composition in dewatered sludge in Japan

2.1 Introduction

Urban development has led to increased construction of sewers worldwide, and an associated increase in the production of sewage sludge [1], an abundant by-product of wastewater treatment plants (WWTPs). After being metabolized by the human body, sewage waste is circulated widely in the environment, and it is rich in inorganic matter, organic matter, and heavy metals [2]. It should therefore be recycled, treated, or safely disposed [3]. Sewage sludge is defined as biomass and has carbon neutrality [4]. Given the large and increasing global sludge production, in addition to traditional sewer management frameworks, material recycling and energy recovery of sewage sludge are essential for future sustainable development [5]. Before sewage sludge recycling, the characteristics and composition of the sludge must be grasped first, so that there is a way to optimize the sludge recycling effect.

Many countries have conducted national sludge surveys in order to properly dispose of sludge. Eriksson et. al [6] investigated 61 trace elements in the sewage sludge of 50 WWTPs in Sweden; Vrien et. al [7] accessed the concentration of 69 elements in the sewage sludge from 64 WWTPs in Switzerland. The United States Environmental Protection Agency (U.S. EPA) investigated 66 sewage sludge samples from 74 WWTPs in 36 states in the U.S. and Venkatesan et. al [8] also collected sewage sludge from 74 WWTPs and analyzed 28 metals.

The chlorine (Cl) in sludge may originate from chlorinated organic compounds or microplastics and should be addressed during recycling [9]. During agricultural recycling, the microplastics in sewage sludge are retained for a much longer time than nutrients in the soil, which poses a threat to the soil ecosystem [10]. Although microplastics do have lethal effects, long-term exposure is considered a key issue due to chronic toxicity [11]. In the cement industry, the Cl content should also be limited when sludge is recycled [12]. However, investigations of Cl were not mentioned in the national surveys.

This study aims to investigate the elemental composition of sewage sludge from 32 WWTPs at the national level in Japan and elucidate influential factors on the elemental composition through statistical analyses. Representative elemental compositions for sewage sludge concerning different parameters are also confirmed through these factors.

2.2 Materials and methods

2.2.1 Materials

Dewatered sludge was collected from 32 WWTPs, chosen to be representative of the following parameters: (1) sewage collection system (combined collection or separated collection), (2) digestion process (digested or undigested), (3) the coagulant used for sludge dewatering (polymer only, polymer + polyferric sulfate, or polyaluminum chloride), (4) dewatering device (centrifuge, belt press, screw press, multi-disk, or rotary press), and (5) geographical location

in Japan. Dewatered sludges from each WWTP were sampled in four seasons over 1 year. The sampling periods were September 9–18, 2015 (summer), October 30 to December 1, 2015 (autumn), February 29 to March 11, 2016 (winter), and May 9–29, 2016 (spring). However, not all WWTPs were sampled in each season due to plant maintenance during some of these periods. Table 2.1 shows the details of the 119 samples that were collected in this study and considered to be fully representative of dewatered sewage sludge in Japan.

Table 2.1 Number and details of samples used in this study.

Dewatering device	Sewage collection	Digestion process	Number of samples	Sampling season			
				Summer	Autumn	Winter	Spring
Centrifuge	Combined	Undigested	1	1	1	1	1
		Digested	2(1)[a]	2	2	2	1
	Separated	Undigested	2(1)[a]	2	2	2	2
		Digested	2(1)[a]	2	1	1	1
Screw press	Combined	Undigested	3(2)[a]	3	3	2	2
	Separated	Undigested	2(1)[a]	2	2	2	2
		Digested	2(1)[a]	2	2	2	2
Multi-disk	Combined	Digested	1	1	1	1	1
	Separated	Undigested	3(2)[a]	3	3	3	3
		Digested	1	1	1	1	1
Belt press	Combined	Undigested	4(2)[a]	2	3	2	2
		Digested	1	1	1	1	1
	Separated	Undigested	1	2	2	2	2
		Digested	2(1)[a]	2	2	2	2
Rotary press	Combined	Undigested	2(1)[a]	2	2	2	2
	Separated	Undigested	2(1)[a]	2	2	2	2
		Digested	1	1	1	1	1
Total			32(14)[a]	31	31	29	28

[a] Number in parentheses shows the number of two coagulants (polymer + polyferric sulfate or polyaluminum chloride) for sludge dewatering

2.2.2 Experimental method

After sampling, the dewatered sludge was placed in a refrigerator in the dark and maintained at or below 4°C. Proximate analysis of samples, specifically moisture content and volatile matter, was conducted according to the Japan Industrial Standard [13][14]. Ultimate analysis was conducted on dried samples using a CHN analyzer (JM-10; J-Science Lab Co. Ltd., Kyoto, Japan) [15], and an X-ray fluorescence spectrometer (XRF; XRF-1800; Shimadzu Co. Ltd., Kyoto, Japan). The CHN analyzer measures the percentages of carbon (C), hydrogen (H), and nitrogen (N) on a dry basis. XRF was used to measure the percentages of main elements including sulfur (S), aluminum (Al), calcium (Ca), iron (Fe), potassium (K), magnesium (Mg), boron (B), barium (Ba), P, silicon (Si), and titanium (Ti) on a dry basis. Volatile matter (V) consists of C, H, N, combustible S, and oxygen (O); however, the XRF analyzer could not measure the O percentage accurately, so the percentage of O was calculated using Eq. (2.1).

$$O = V - C - H - N - Sc \qquad (2.1)$$

Where Sc indicates combustible S, which was assumed to equal to total S (Appendix, A1).
The heating value of sludge was determined using an adiabatic bomb calorimeter (CA-4J; Shimadzu Co. Ltd.). The lower heating value (LHV) of dried and dewatered samples was calculated from the measured higher heating values (HHV) using Eqs. (2.2) and (2.3)

$$LHV_d = HHV - 4.186 \times 600 \times 9 \times H \qquad (2.2)$$

$$LHV_w = \frac{100-w}{100} \times LHVd - 4.186 \times 600 \times \left(\frac{w}{100}\right) \qquad (2.3)$$

Where LHV_d and LHV_w represent the LHV of the dried and dewatered samples, respectively, w is the moisture content of the dewatered samples, and H (% dry basis) is the hydrogen concentration of the sludge.

Heavy metals in sewage sludge, including cadmium (Cd), chromium (Cr), copper (Cu), manganese (Mn), sodium (Na), nickel (Ni), lead (Pb), and zinc (Zn) were analyzed after acid digestion. A dry sludge sample weighing 0.5 g was placed in a 100-mL Erlenmeyer flask. HNO_3 (69%, SP; Nacalai Tesque, Inc., Kyoto, Japan) and HCl (35%, SP; Nacalai Tesque, Inc., Kyoto, Japan) were added in a volume ratio of 1:1. The mixture was then heated on a hot plate to almost dryness and HNO_3 and HCl were added in the same ratio. The above procedures were repeated until the liquid became light yellow and the precipitate was grayish. Metal concentrations were analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES) (ICPS-8000; Shimadzu Co. Ltd., Kyoto, Japan). All analyses were conducted in triplicate and the average was taken.

According to the literature, neutron activation analysis (NAA) can provide good quantification of low-concentration elements without matrix effects and can be used to analyze various elements simultaneously [16]. The bromine (Br) and Cl contents of the samples were analyzed by NAA using the Kyoto University Research Reactor (KUR), Pn-3 (neutron beam of about $2.0–2.4 \times 10^{13} \, cm^{-2} \, S^{-1}$). The radionuclides ^{38}Cl ($t_{1/2} = 37.18$ min, $E_\gamma = 1{,}642$ keV) and ^{80}Br ($t_{1/2} = 17.6$ min, $E_\gamma = 617$ keV) were generated by irradiation for 10 min, and then measured using a germanium semiconductor detector for 5 min. The comparison method was used for quantification, in which a standard with a known content of the target element is subjected to neutron emission γ-ray measurement under the same conditions as the sample, and the peaks are compared to determine the concentration of the target element in the sample. A 1-cm square filter paper (No.5C; Advantec) and a mixed aqueous solution of NH_4Cl and NH_4Br (Wako Pure Chemical Industries, Ltd., Osaka, Japan) were used as standard samples, and a quantitative comparison of the results was conducted using the NZMCA analysis software (ver. 1.0.1.6; Laboratory Equipment Corp.).

2.2.3 Quality control of ICP-AES result

Quality control during the ICP-AES analysis was conducted using the BCR-176 municipal solid waste standard. BCR-176 is incineration fly ash that was collected in Brussels in November 1984, which the Commission of the European Communities markets adopted for use as a standard for recovery of various elements in ICP-AES analysis including acid digestion pre-treatment. Takaoka et al. [17] compared the accuracy of three different methods for BCR samples and proposed the BCR elemental composition measured by ICP-AES as a certified value. Table 2.2 shows the measured value as an average of triplicate measurements and the reference value. The analysis confirmed recoveries in the range of 80–120% for the elements Cd, Cr, Cu, Mn, Na, Ni, Pb, and Zn, which was within the acceptable variation of 70–130% recommended by the U.S EPA quality control standard [18].

Table 2.2 Certified reference values and measured values for reference samples used during the analysis of sludge by inductively coupled plasma atomic emission spectrometry. Sample ID: BCR-176.

Element	Reference value[17] (mg/kg DS)	Value measured[a] (mg/kg DS)	Recovery ratio[b]
Cd	470	546	116%
Cr	863	733	85%
Cu	1300	1310	101%
Mn	1400	1420	101%
Na	31000	30100	97%
Ni	123	125	102%
Pb	10800	12500	115%
Ti	10000	342	3%
Zn	25700	26200	102%

[a] Average of triplicate results [b] Ratio of the measured value to the reference value

2.3 Results and discussion

2.3.1 Element analysis

Table 2.3 shows the average sludge compositions from the 32 WWTPs. The moisture contents of the dewatered sludge samples ranged from 72.1% to 85.7% (average: 79.8%), and the volatile matter from 64.6% to 88.7% (average: 80% dry basis). From the total arithmetic mean, the element concentrations were in the order C > O > H > N > P > Si > Fe > Ca > Al > S > Mg > K > Cl > Ti > Na > Ba > Zn > Mn > Cu > Br > Cr > Ni > Pb > Cd. This tendency for heavy metal concentrations, where Zn > Cu > Cr > Ni > Cd, was consistent with previous studies on sewage sludge [19][20].

Table 2.3 Composition of the sewage sludge from 32 wastewater treatment plants in Japan.

No	Sewage [a] collection	Dewatering	Digestion	Coagulants [b]	M [c] (%)	V [d] (%)	HHV (kJ/kg-DS)	LHVd [e] (kJ/kg-DS)	LHVw [f] (kJ/kg)	Element (mg/kg-DS)									Note
										C	O	H	N	P	Si	Fe	Ca	S	
1	Com.	Centrifuge	Undigested	Single	79.4	82.2	18100	16700	1430	407000	304000	65400	36100	18100	26300	22900	12200	9240	
2	Com.	Centrifuge	Digested	Single	80.5	64.6	14700	13500	600	322000	198000	55600	48800	40400	32300	53500	20500	21000	
3	Com.	Centrifuge	Digested	Double	80.4	65.2	15100	13800	694	322000	210000	55200	45900	39200	37200	48000	20300	18400	not in spring
4	Sep.	Centrifuge	Undigested	Single	84.0	87.6	20000	18400	845	440000	269000	70900	81900	30700	13100	10000	13200	13400	
5	Sep.	Centrifuge	Undigested	Double	80.1	84.4	19600	18100	1590	429000	267000	69400	68400	28400	12500	16500	8360	10400	
6	Sep.	Centrifuge	Digested	Single	85.5	74.3	17500	16000	186	380000	227000	62800	55300	31600	28200	11800	23200	18200	summer only
7	Sep.	Centrifuge	Digested	Double	79.2	73.8	16300	14900	1110	354000	247000	61000	61500	37100	17800	20700	19800	14200	
8	Com.	Screw press	Undigested	Single	79.6	78.6	17700	16300	1310	395000	272000	62100	47500	27600	29500	16000	11400	9640	
9	Com.	Screw press	Undigested	Double	77.6	81.6	18300	16900	1840	409000	277000	64700	53700	23500	23700	19400	8370	11400	summer and autumn only
10	Com.	Screw press	Undigested	Double	76.2	85.8	18900	17300	2220	429000	300000	68700	48200	20200	19100	14600	8900	11200	
11	Sep.	Screw press	Undigested	Single	78.4	88.7	19700	18000	1940	441000	311000	71300	55400	21800	14200	6490	11100	8090	
12	Sep.	Screw press	Undigested	Double	81.8	84.1	18700	17200	1070	417000	288000	67400	55400	24300	13900	13800	10700	13100	
13	Sep.	Screw press	Digested	Single	85.7	76.2	17700	16200	155	383000	237000	65300	59000	30000	23000	18900	19400	17100	
14	Sep.	Screw press	Digested	Double	82.5	74.1	17000	15600	662	367000	234000	63200	60000	41400	18500	35500	20200	17000	
15	Com.	Multi desk	Digested	Single	78.4	76.8	17900	16500	1600	384000	239000	63900	62100	27000	28300	26500	16100	19600	
16	Sep.	Multi desk	Undigested	Single	83.4	79.1	18200	16700	671	392000	250000	67900	71500	28600	25100	4350	10600	9030	
17	Sep.	Multi desk	Undigested	Double	85.2	87.0	20000	18300	566	432000	268000	71800	84300	24100	6280	24300	5370	14300	
18	Sep.	Multi desk	Undigested	Double	81.9	82.5	18200	16700	969	401000	273000	67400	71300	26300	23200	18500	6590	12300	
19	Sep.	Multi desk	Digested	Single	79.2	68.0	15200	13900	909	334000	226000	56600	50100	36300	31200	55000	13300	13500	
20	Com.	Belt press	Undigested	Single	77.5	86.8	19800	18200	2150	437000	296000	68600	56700	24300	10100	13000	7610	9600	autumn only
21	Sep.	Belt press	Undigested	Single	79.0	87.4	19100	17500	1700	429000	319000	69500	49300	18900	17700	4860	12500	7550	
22	Com.	Belt press	Undigested	Double	79.3	79.3	18100	16600	1440	397000	268000	63900	52600	24300	24200	28800	13000	11600	
23	Com.	Belt press	Undigested	Double	77.5	81.3	18300	16800	1840	406000	273000	65700	56200	27300	20800	25300	7960	12400	
24	Com.	Belt press	Digested	Single	84.3	73.3	17200	15800	349	372000	224000	61200	60200	32700	35000	21000	21000	16200	
25	Sep.	Belt press	Undigested	Single	75.9	87.6	19400	17800	2390	435000	312000	69800	50800	21100	13800	4020	10200	8140	
26	Sep.	Belt press	Digested	Single	83.2	81.5	19100	17600	861	415000	246000	67500	73100	30700	22700	6500	17900	12400	
27	Sep.	Belt press	Digested	Double	77.2	71.2	15400	14100	1280	332000	238000	58200	56300	39000	16200	57900	9350	27200	
28	Com.	Rotary press	Undigested	Single	73.2	78.8	17800	16400	2510	400000	272000	63600	43100	18900	33800	15400	16500	9420	
29	Com.	Rotary press	Undigested	Double	76.2	78.0	17900	16500	2000	386000	279000	62400	42900	24800	27600	36000	9220	8930	
30	Sep.	Rotary press	Undigested	Single	72.1	88.1	19400	17900	3180	440000	321000	69600	43400	16900	16600	3980	9610	7640	
31	Sep.	Rotary press	Undigested	Double	77.2	85.5	18700	17200	1980	417000	312000	68000	49000	22600	11700	30100	6270	8630	
32	Sep.	Rotary press	Digested	Single	82.5	85.0	20100	18400	1150	434000	265000	71100	66100	22200	18700	4760	18900	13400	
	Average				79.8	79.9	18100	16600	1350	398000	266000	65300	56700	27500	21600	21500	13100	12900	
	Median				79.4	81.4	18200	16700	1290	403000	269000	65500	55400	26700	21800	18700	11800	12400	
	Standard deviation				3.42	6.75	1480	1380	727	35600	32900	4610	11300	6880	7870	15200	5150	4530	

[a] Com: combined; Sep: separated. [b] Single: polymer; Double: polymer + polyferric sulfate or polyaluminum chloride. [c] M: moisture content.

[d] V: volatile matter. [e] LHVd: LHV of dried sample [f] LHVw: LHV of dewatered sample

Table 2.3 cont.

No	Sewage [a] collection	Dewatering	Digestion	Coagulants [b]	Element (mg/kg-DS)															Note
					Al	Mg	K	Cl	Ti	Ba	Na	Zn	Cu	Mn	Br	Ni	Pb	Cr	Cd	
1	Com.	Centrifuge	Undigested	Single	7780	4070	3440	1560	765	586	690	630	130	734	95.7	11	26.2	6.4	1.25	
2	Com.	Centrifuge	Digested	Single	15300	11500	3850	940	1430	1050	1930	1490	665	950	70.1	129	55.6	99.5	1.51	
3	Com.	Centrifuge	Digested	Double	20300	7140	3860	1340	1780	778	1140	2180	813	984	63.4	427	64.7	426	1.54	not in spring
4	Sep.	Centrifuge	Undigested	Single	10500	11100	4400	1070	742	553	3010	207	234	69.3	87.6	17.3	5.43	4.47	0.62	
5	Sep.	Centrifuge	Undigested	Double	7230	4590	3630	1030	764	797	328	420	240	117	113	11	8.62	4.06	0.552	
6	Sep.	Centrifuge	Digested	Single	19500	4070	1880	1310	1280	909	644	798	598	241	60.8	17.4	22.4	9.4	1.78	summer only
7	Sep.	Centrifuge	Digested	Double	12300	12300	3200	828	857	736	961	454	253	758	31.1	18.8	18	18.8	1.4	
8	Com.	Screw press	Undigested	Single	21600	7170	4010	1040	1120	564	539	458	241	525	71.9	18.3	32.1	23.8	1.29	
9	Com.	Screw press	Undigested	Double	13500	3840	3530	950	1010	660	662	611	293	708	50.9	29.4	23.2	38.9	0.647	summer and autumn only
10	Com.	Screw press	Undigested	Double	11800	3830	3280	1550	855	655	873	437	170	124	98.3	23.5	21.2	68.6	0.655	
11	Sep.	Screw press	Undigested	Single	8150	4890	3480	993	589	698	573	455	212	184	61.8	68	6.55	7.15	0.718	
12	Sep.	Screw press	Undigested	Double	11000	4750	4050	1300	708	709	1790	685	323	438	50.5	51.1	17.7	27.2	1.27	
13	Sep.	Screw press	Digested	Single	16800	5310	2750	1910	1470	1440	643	753	615	260	99.9	16.9	23.3	8.25	1.52	
14	Sep.	Screw press	Digested	Double	9590	11800	3480	1080	1220	973	637	584	1120	310	150	53.3	21.7	25.9	0.831	
15	Com.	Multi desk	Digested	Single	12000	8990	3040	1210	1430	1610	2420	654	356	147	40.2	19.4	24.6	10.7	1.13	
16	Sep.	Multi desk	Undigested	Single	33700	4290	4240	1130	630	450	611	283	154	95.1	50.5	14.6	7.74	2.53	0.922	
17	Sep.	Multi desk	Undigested	Double	3330	4170	5580	905	679	359	733	329	186	135	79.7	30.2	5.23	8.3	0.531	
18	Sep.	Multi desk	Undigested	Double	15600	2810	4280	1700	873	1050	326	443	572	210	69.1	16.2	11.3	4.33	0.71	
19	Sep.	Multi desk	Digested	Single	16600	5860	2730	994	1300	1410	621	651	619	178	60	40	27	94.5	0.774	
20	Com.	Belt press	Undigested	Single	5910	5110	2930	2400	771	576	743	306	277	223	57.5	8.12	12.8	3.5	0.525	autumn only
21	Sep.	Belt press	Undigested	Single	7790	4840	4680	1390	874	522	495	392	171	71.8	73.7	15.6	11.8	3.33	0.709	
22	Com.	Belt press	Undigested	Double	13700	3460	3510	1130	1490	647	625	1190	462	607	122	70.8	19.7	259	0.816	
23	Com.	Belt press	Undigested	Double	9150	4450	4130	856	927	539	507	686	284	739	24.7	33	19.3	59	0.597	
24	Com.	Belt press	Digested	Single	16700	9940	3720	948	1540	1320	857	1070	534	759	38.4	127	37	109	1.69	
25	Sep.	Belt press	Undigested	Single	11200	4980	3320	870	582	623	432	1950	249	261	33.1	14.8	6.42	13.9	0.687	
26	Sep.	Belt press	Digested	Single	11400	9090	4030	1490	1100	955	645	410	304	443	84.4	35	12.5	12.8	1.08	
27	Sep.	Belt press	Digested	Double	8620	9400	2080	951	1180	735	995	478	268	192	41.7	37.4	18.9	19.1	0.629	
28	Com.	Rotary press	Undigested	Single	11200	6110	4150	1590	1100	584	1010	501	186	174	90.7	15.4	17	8.02	0.678	
29	Com.	Rotary press	Undigested	Double	9670	5260	3700	1010	820	697	509	578	144	1630	138	12.5	25.2	5.87	1.16	
30	Sep.	Rotary press	Undigested	Single	5930	4610	2420	946	620	449	501	257	137	181	64.7	6.95	7.56	1.49	0.874	
31	Sep.	Rotary press	Undigested	Double	5900	2680	2970	1090	621	914	330	343	163	88.6	48.9	15.5	8.92	32.2	0.645	
32	Sep.	Rotary press	Digested	Single	9350	5450	2680	798	1030	2220	757	537	426	158	37.1	11.3	14.1	5.32	1.35	
	Average				12300	6180	3530	1200	1000	836	860	663	356	397	70.6	44.2	19.8	44.4	0.972	
	Median				11200	5050	3520	1080	900	703	644	519	273	232	64.1	18.6	18.5	11.8	0.823	
	Standard deviation				5910	2790	768	355	326	396	606	457	229	356.0	31.0	76.0	13.3	86.2	0.38	

[a] Com: combined; Sep: separated. [b] Single: polymer; Double: polymer + polyferric sulfate or polyaluminum chloride

The elemental concentration distribution of the 119 samples is shown in Figure 2.1. The concentrations of most elements were extremely high. Moreover, the arithmetic means of Ca, Cl, Na, Ba, Zn, Cu, Mn, Br, Cr, Ni, and Pb were 10% higher than the median, indicating that higher values affected the overall concentration distribution. In particular, for Ni and Cr, the maximum was about 10 times the arithmetic mean.
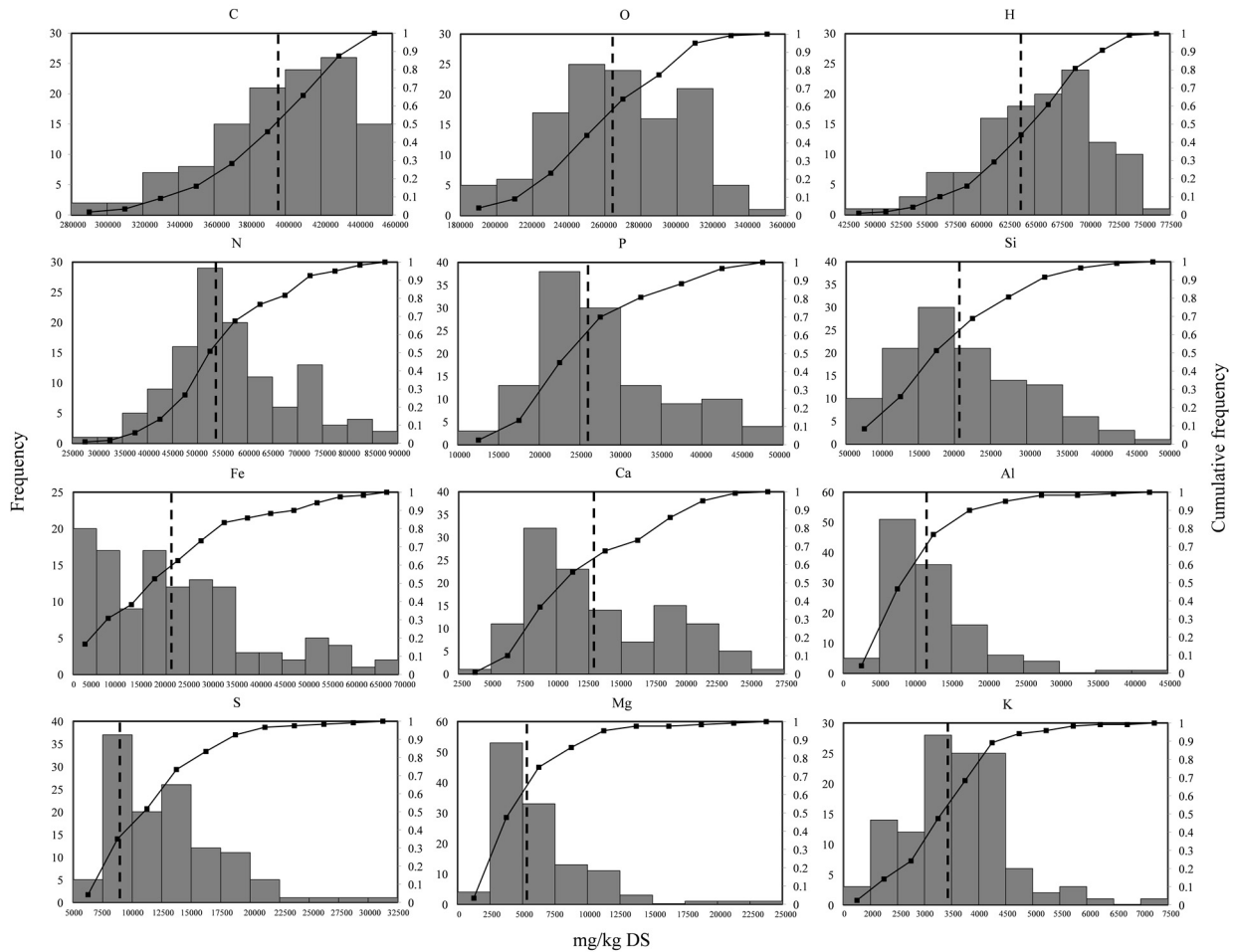
Figure 2.1 Elemental concentration distribution of the elements in sludge from the wastewater treatment plant. Left axis, the frequency for each sample; right axis, cumulative frequency; horizontal axis, range of elemental concentrations; black dotted line, arithmetic mean; red line, Japanese regulatory standard for heavy metals in sludge for agricultural use.
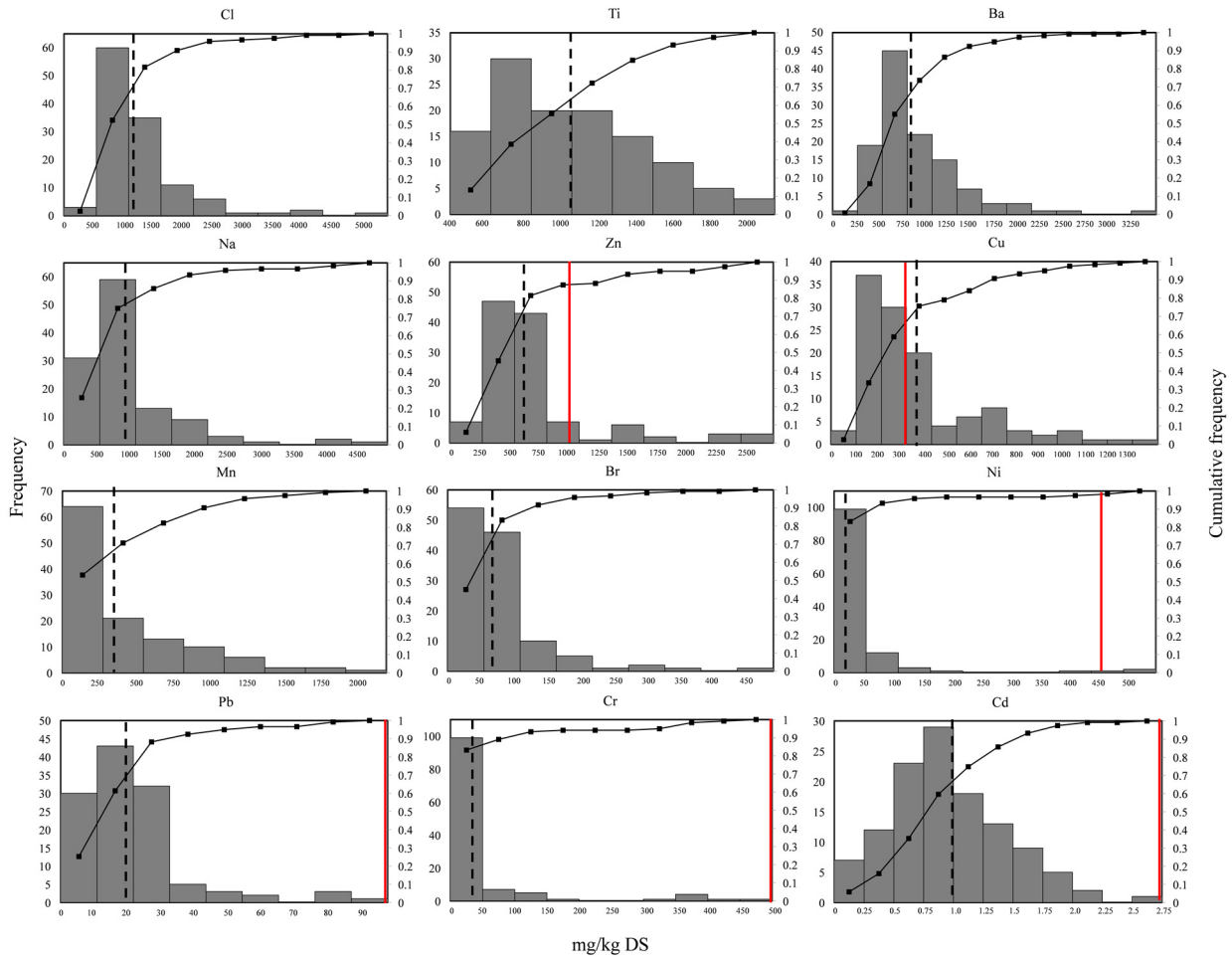
Figure 2.1 cont.

For the distributions of the elemental concentrations, the normal and log-normal verification results for each element are shown in Table 2.4 and Figures A2.1-A2.3 in the Appendix. The Shapiro–Wilk Normality Test was applied to verify that the data followed a normal distribution, where smaller values under Prob < W indicate that the data are less likely to follow a normal distribution and values greater than 0.05 indicate a normal distribution. Kolmogorov's D statistic was used to determine whether the data had a log-normal distribution, where Prob>D values greater than 0.05 indicate that the data is likely to have a log-normal distribution. Skewness refers to asymmetry in a curve, where a negative (positive) value of skewness indicates that the overall data has a negatively (positively) skewed distribution. From these analyses, O and H were normally distributed, whereas N, P, Si, Al, B, and Mn were log-normally distributed. C was negatively skewed, and all other elements (S, Fe, Ca, Mg, K, Cl, Ti, Na, Zn, Cu, Br, Ni, Pb, Cr, and Cd) were positively skewed. Finally, S, Fe, Ca, and Cu exhibited bimodal distributions.

Table 2.4 Distribution analysis of elements in three countries.

Element	Japan (N=119)						Sweden (N=50) [6]						Switzerland (N=64) [7]					
	Normal distribution		Log normal		Skewness	Judgment	Normal distribution		Log normal		Skewness	Judgment	Normal distribution		Log normal		Skewness	Judgment
	Shapiro-Wilk Test		Kolmogorov's D				Shapiro-Wilk Test		Kolmogorov's D				Shapiro-Wilk Test		Kolmogorov's D			
	W	Prob<W	D	Prob>D			W	Prob<W	D	Prob>D			W	Prob<W	D	Prob>D		
C	0.939	<.0001	0.112	<0.01	-0.769	Negative skewed												
O	0.984	0.152	0.064	>0.15	-0.128	Normal												
H	0.981	0.084	0.077	0.078	-0.475	Normal												
N	0.974	0.0211	0.073	0.117	0.466	Log normal												
S	0.914	<.0001	0.082	0.048	1.16	Bimodal	0.884	0.0001	0.354	<0.01	1.49	Positive skewed	0.543	<.0001	0.196	<0.01	4.98	Positive skewed
P	0.944	<.0001	0.072	0.125	0.756	Log normal	0.973	0.312	0.361	<0.01	-0.135	Normal	0.965	0.065	0.105	0.0791	0.37	Bimodal
Si	0.958	0.0009	0.051	>0.15	0.713	Log normal	0.850	<.0001	0.262	<0.01	1.80	Positive skewed	0.850	<.0001	0.277	<0.01	0.38	Positive skewed
Fe	0.911	<.0001	0.116	<0.01	0.857	Bimodal	0.895	0.0004	0.164	<0.01	0.638	Bimodal	0.980	0.367	0.166	<0.01	0.19	Normal
Ca	0.930	<.0001	0.106	<0.01	0.521	Bimodal	0.528	<.0001	0.290	<0.01	3.60	Positive skewed	0.989	0.835	0.154	<0.01	0.01	Normal
Al	0.872	<.0001	0.049	>0.15	1.66	Log normal	0.918	0.002	0.219	<0.01	0.30	Bimodal	0.731	<.0001	0.125	0.020	1.93	Bimodal
Mg	0.793	<.0001	0.103	<0.01	2.12	Positive skewed	0.975	0.378	0.361	<0.01	-0.19	Normal	0.773	<.0001	0.241	<0.01	2.24	Positive skewed
K	0.960	0.0013	0.092	0.018	0.69	Positive skewed	0.903	0.001	0.300	<0.01	1.18	Positive skewed	0.887	<.0001	0.178	<0.01	1.28	Positive skewed
Cl	0.763	<.0001	0.098	<0.01	2.43	Positive skewed												
Ti	0.943	<.0001	0.091	0.023	0.598	Positive skewed							0.980	0.373	0.073	>0.15	0.02	Log normal
Ba	0.832	<.0001	0.073	0.121	2.06	Log normal	0.949	0.035	0.120	0.078	0.64	Log normal	0.978	0.292	0.078	>0.15	0.51	Log normal
Na	0.684	<.0001	0.129	<0.01	2.78	Positive skewed	0.403	<.0001	0.175	<0.01	5.71	Positive skewed	0.619	<.0001	0.189	<0.01	3.13	Positive skewed
Zn	0.702	<.0001	0.125	<0.01	2.32	Positive skewed	0.618	<.0001	0.128	0.0433	3.97	Positive skewed	0.934	0.002	0.060	>0.15	1.19	Log normal
Cu	0.804	<.0001	0.103	<0.01	1.69	Bimodal	0.714	<.0001	0.083	>0.15	2.97	Log normal	0.528	<.0001	0.143	<0.01	4.86	Positive skewed
Mn	0.795	<.0001	0.078	0.077	1.71	Log normal	0.782	<.0001	0.086	>0.15	1.99	Log normal	0.890	<.0001	0.060	>0.15	1.39	Log normal
Br	0.720	<.0001	-	-	2.87	Positive skewed												
Ni	0.407	<.0001	0.141	<0.01	4.44	Positive skewed	0.386	<.0001	0.088	>0.15	5.99	Log normal	0.613	<.0001	0.143	<0.01	3.38	Positive skewed
Pb	0.796	<.0001	0.082	0.0495	2.17	Positive skewed	0.805	<.0001	0.087	>0.15	2.51	Log normal	0.583	<.0001	0.135	<0.01	4.61	Positive skewed
Cr	0.469	<.0001	-	-	3.33	Positive skewed	0.956	0.060	0.090	>0.15	0.52	Normal	0.679	<.0001	0.099	0.144	3.59	Log normal
Cd	0.978	0.049	-	-	0.602	Positive skewed	0.339	<.0001	0.135	0.0314	6.30	Positive skewed	0.561	<.0001	0.373	<0.01	2.80	Positive skewed

* Gray cells: p > 0.05

In Sweden (N = 50), P, Mg, and Cr were normally distributed; Ba, Cu, Mn, Ni, Pb, and Cr followed a log-normal distribution; S, Si, Fe, Ca, Al, K, Na, Zn, and Cd were positively skewed; and Al and Fe had bimodal distributions [6]. In Switzerland (N = 64), P, Fe, and Ca were normally distributed; Ti, Ba, Zn, Mn, and Cr followed a log-normal distribution; S, Si, Al, Mg, K, Na, Cu, Bi, Pb, and Cd were positively skewed; and P and Al had bimodal distributions [7]. Among the values obtained for these three countries, only Ba and Mn consistently followed a log-normal distribution, and most elements were positively skewed, indicating that the arithmetic mean of the element concentration was greater than the median. The distribution of most heavy metals followed a log-normal distribution in Sweden, but no such trend was observed in Japan and Switzerland. A bimodal distribution was seen for S, P, Al, and Fe in all three countries. Al and Fe salts are used in water treatment processes and in the dewatering of sludge to remove P and increase dehydration; therefore, these distributions may be related to the coagulants used in WWTPs.

Many countries have regulatory standards for heavy metal concentrations when sewage sludge is used in agriculture. Table 2.5 shows national standards for sludge for agricultural use, including those from the United States [21], European Union, Sweden [22], China [23], and Japan [24]. The limits provided differ significantly among countries. The heavy metal concentrations in sludge in this study fell within the United States and European Union regulatory limits; however, the concentrations of Cu and Zn exceeded China's limits. According to the Japanese standard, about 10% of the 119 sludge samples contained excess Zn, 40% contained excess Cu, 4% contained excess Ni. The limits in Sweden were much lower, and the elemental concentrations in all 119 samples were above the Swedish standard. From the above results, Cu is the major contaminant in sewage sludge in Japan when considering agricultural use, where its concentration limit is set to 300 mg/kg-TS.

Table 2.5 Permissible standard limits for sewage sludge used in agriculture.

Heavy metals	Permissible standard (mg/kg)						This study
	United States	European Union	Sweden	China		Japan	
	U.S.EPA-40 CFR 503.13[21]	Directive 86/278/EEC[22]	Government of Sweden[22]	Pollutants control standard GB4284-84[23]		Fertilizer Regulation Act in Japan[24]	Arithmetic mean
				pH < 6.5	pH > 6.5		
Cd	39	20-40	0.75	5	20	5	0.972
Cr	1,200	-	40	600	1,000	500	44.4
Cu	1,500	1,000-1,750	300	250	3	300	356
Pb	300	750-1,200	25	300	1,000	100	19.8
Ni	420	300-400	25	100	200	300	44.2
Zn	2,800	2,500-4,000	600	100	200	900	663

The sludge compositions from various countries are shown in Figure 2.2. The medians and the standard deviations of this study were compared with past survey data from the United States [25][26][27], Sweden [6], Switzerland [7], Japan [28]. The concentrations of the elements H, P, Si, Fe, Ca, Al, Na, Mn, Ni, Pb, Cr, and Cd were highly variable; however, when taking considering the media, all countries exhibited similar trends, indicating that although individual concentrations had extreme values, the overall concentrations of constituent elements in sludges from different countries were similar.
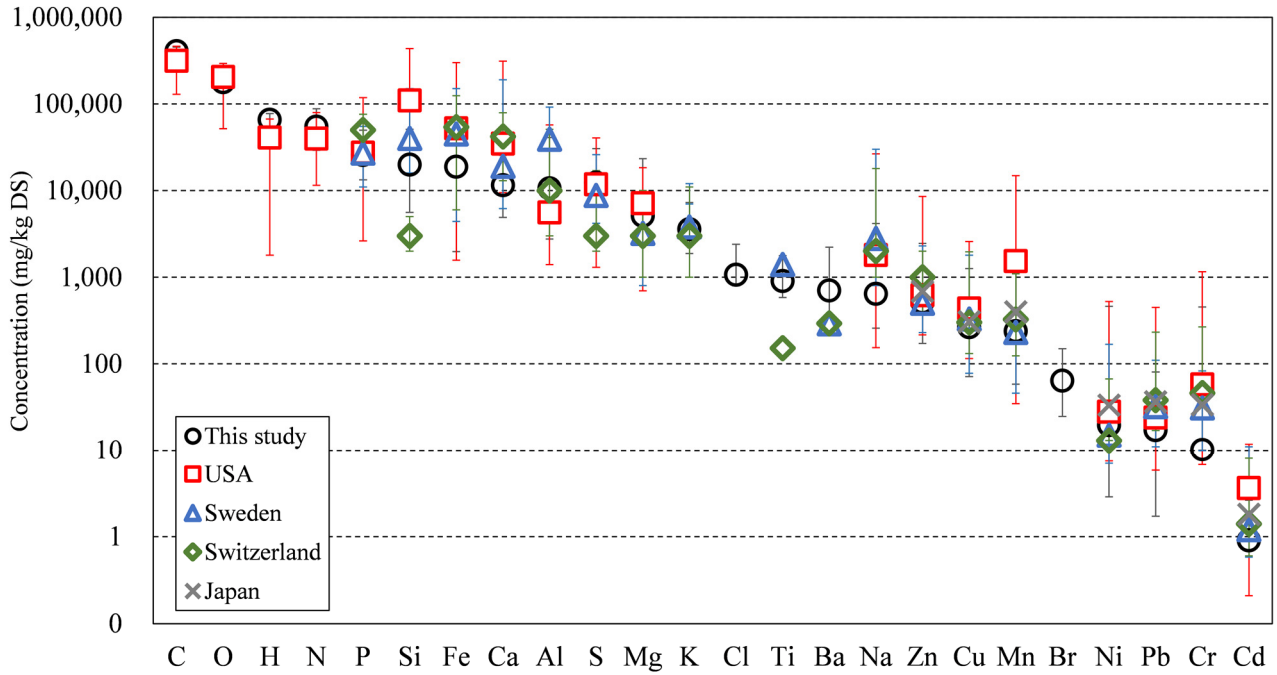
Figure 2.2 Comparison of the composition of sewage sludge from various countries. Error bars show the standard deviation [6][7][25][26][27][28].

2.3.2 Factors affecting sludge properties based on Hayashi's quantification method I

Differences in elemental concentrations in sewage sludge arise mainly due to peoples' living habits, operational processes in WWTPs, or differences in geographical areas covered by urban sewage networks. The elemental composition of dewatered sludge may be influenced by season, sewage collection system, dewatering device, digestion process, and coagulant for dewatering. It is useful to clarify the level of influence of these factors and categorize the elemental compositions according to the factors with the greatest influence. The influence of season, sewage collection system, dewatering device, digestion process, and coagulant used for dewatering on elemental composition in sludge was analyzed using Hayashi's quantification method I [29], which is a multivariate analysis method where the objective variable is quantitative, and the explanatory variables are qualitative. Hayashi's quantification method I can be written as Eq. (2.4).

$$y = (a_{11}x_{11} + a_{12}x_{12} + a_{13}x_{13} + a_{14}x_{14}) + (a_{21}x_{21} + a_{22}x_{22})$$

$$+ (a_{31}x_{31} + a_{32}x_{32} + a_{33}x_{33} + a_{34}x_{34} + a_{35}x_{35})$$

$$+ (a_{41}x_{41} + a_{42}x_{42}) + (a_{51}x_{51} + a_{52}x_{52}) + a_0 \qquad (2.4)$$

where y is the predicted elemental concentration in sewage sludge of each element (mg/kg dry basis); $x_{11} \sim x_{52}$ denote the data quantity and can be 1 or 0; $a_{11} \sim a_{14}$ denote the category scores for the season; $a_{21} \sim a_{22}$ denote the category scores for the sewage collection system; $a_{31} \sim a_{35}$ denote the

category scores for dewatering device; $a_{41}\sim a_{42}$ denote the category scores for digestion process; $a_{51}\sim a_{55}$ denote the category scores for the coagulant used for dewatering; and a_0 is constant.

For the analysis, the qualitative data for each category (factor) were first converted into quantitative data (1 or 0). In this study, the five parameters of the season (four categories), sewage collection system (two categories), dewatering device (five categories), digestion process (two categories), and the coagulant used for dewatering (two categories) were the explanatory variables (x), and the objective variable (y) was the concentration of the elements in the dewatered sludge. The JMP (ver. 14.0; SAS Institute Inc., Cary, NC, USA) and College Analysis statistical analysis software were used for Hayashi's quantification method I.

Table 2.6 shows the estimated influence of each factor on the elemental concentrations in sewage sludge. The multiple correlation coefficient adjusted for the degrees of freedom (R) indicates the estimation accuracy for all categories. The values in Table 2.6 are expressed as a calculated category score divided by the median of each elemental concentration as a percentage, which allows a comparison of the values. In terms of category scores, positive (negative) values indicate an increase (decrease) in concentration. Among the scores in each category, the black cells indicate a significant ($p < 0.01$) correlation between the element and the influential factor. The range is the value of the maximum minus the minimum value of the parameter, where larger ranges indicate a greater impact on the element, which allows a comparison of the impact of each factor on the same element.

Br, Cl, Na, K, Zn, Cr, Ni, and Al had low R values (< 0.5), indicative of relatively low influences on these elements. The alkali metals, Na and K, and halogens, Cl and Br, easily form alkali halides and dissolve in water [30]; therefore, it is assumed that these elements preferentially flow with the separated water during the dewatering process, with less remaining in the dewatered sludge, resulting in the lower correlation between their concentrations and the categories. Metals such as Zn, Cr, Ni, and Al are more commonly found in industrial wastewater than in domestic wastewater, which was likely the dominant influence on these elements, rather than sewage treatment conditions. The other elements had higher R values (> 0.5), indicating that the categories could explain these elemental concentrations to some extent. Organic elements such as C, O, H, N, and S, and the volatile matter had higher R values than Ca, P, Si, Ba, Ti, and Pb, indicating that these elements were more susceptible to the categories.

Overall, the sewage collection system, digestion process, and coagulants used for dewatering had a greater impact on elemental concentrations, while season and dewatering devices had minimal effects. The organic elements C, O, H, N, and S, as well as volatile matter, were affected by the sludge collection system, digestion process, and coagulant used for dewatering, although the coagulant used for dewatering had a much smaller impact. Considering the sewage collection system and digestion process, the organic matter content tended to be lower in combined and digested systems; however, even though some S in sludge was likely converted into digestion gases such as H_2S, S still tended to be concentrated by the decomposition of other organic materials in dewatered sludge.

Combined sewage collection systems with digestion tended to lead to an increase in P, Si, Fe, Ca, Mg,

Ti, Ba, Cu, Mn, Pb, and Cd, indicating that these elements were concentrated in dewatered sludge compared to natural sources such as rainwater and soil. With the addition of inorganic coagulants, volatile matter and other organic matter tended to decrease. This may be due to a decrease in relative concentrations. However, the opposite effect was found for S, where the concentration tended to increase. One possible explanation is that most of the inorganic coagulant used was in the form of sulfate. The addition of inorganic coagulants had a positive effect on the concentrations of Mn, Fe, Cu, and Cr, because these may be co-precipitated with Fe ions or coagulants. Other elements were not significantly affected by coagulants for dewatering.

2.3.3 Correlation between elements

Table 2.7 shows the single correlation coefficients between various elements. C was positively correlated with O and H. This is expected because C, H, and O are the main elements of organic matter. In addition, Mg and P were significantly correlated. A previous study showed that the Mg concentration can affect the biological uptake behavior of phosphate [31], and Harold [32] confirmed that polyphosphates are involved in many biochemical and physiological processes and metabolic regulation and that Mg can serve as a bridge to form complexes. Therefore, the concentrations of Mg and P likely affect each other. Si and Ti, also correlated, are common elements required for plant growth, which in sewage are affected by fertilizers, soil, rocks, and other natural conditions. The correlation between these elements is likely related to the sewage collection and the possibility of contamination with rainwater or wastewater other than domestic water. Ni and Cr have the advantages of corrosion resistance and moderate cost and are thus often used together in electric heating elements, stainless steel, and other metal materials. The same significant correlation was also observed between Ni and Pb, possibly related to the incorporation of industrial wastewater.

By contrast, C was negatively correlated with Fe, P, and Ti. Regarding Fe and P, iron salts may be added to biological treatments to remove P in some WWTPs. Additionally, iron salt may be used prior to sludge thickening and dewatering to improve the elution of P in the solid phase. In this case, Fe reacts with P to become the insoluble inorganic salt: $FePO_4$ and remains in the dewatered sludge. Therefore, it is speculated that the concentration of C decreased, and is negatively correlated with Fe and P. The reasons for the relationship between C and Ti have not yet been clarified. Regarding the other elements, H was negatively correlated with Pb, and O was negatively correlated with Ti. Possible reasons have not yet been clarified.

Table 2.6 Summary of the influence of factors on the properties of sewage sludge and the concentration of each element based on Hayashi's quantification method I.

%	R[a]	Season					Sewage collection			Dewatering device[b]						Digestion process			Coagulants [c]		
		Autumn	Summer	Winter	Spring	Range	Separated	Combined	Range	SP	BP	RP	DP	CE	Range	Digested	Undigested	Range	1	2	Range
M	0.513	0.276	-0.247	0.083	-0.112	0.52	1.05	-1.05	2.10	1.06	-0.105	-4.04	1.88	1.22	5.92	1.08	-1.08	2.16	0.156	-0.156	0.31
V	0.793	0.732	-2.45	1.77	-0.046	4.22	3.10	-3.10	6.20	1.02	2.21	1.48	-2.57	-2.14	4.78	-6.57	6.57	13.1	1.44	-1.44	2.88
C	0.770	1.75	-2.81	1.60	-0.541	4.56	2.86	-2.86	5.72	1.44	1.79	1.94	-3.25	-1.92	5.19	-6.64	6.64	13.3	2.09	-2.09	4.18
O	0.811	-0.617	-1.47	1.79	0.298	3.26	2.33	-2.33	4.06	1.80	2.74	5.09	-6.32	-3.32	11.4	-9.98	9.98	20.0	1.65	-1.65	3.30
H	0.757	-2.64	-4.17	4.05	2.77	8.22	3.31	-3.31	6.62	0.959	1.29	0.477	-1.08	-1.64	2.93	-4.85	4.85	9.70	1.39	-1.39	2.78
N	0.645	3.69	-3.08	1.28	-1.88	6.77	10.7	-10.7	21.4	-6.00	3.63	-14.8	15.6	1.54	30.4	0.113	-0.113	0.23	-2.05	2.05	4.10
S	0.797	-0.261	5.35	-4.61	-0.477	9.96	-2.79	2.79	5.58	-0.133	2.77	-14.9	8.16	4.14	23.1	29.4	-29.4	58.8	-11.0	11.0	22.0
P	0.722	-1.68	5.72	-5.5	1.45	11.2	1.63	-1.63	3.26	0.402	0.391	-17.1	3.27	13.0	30.1	19.7	-19.7	39.4	-7.35	7.35	14.7
Si	0.700	-10.7	21.5	-12.7	1.95	34.2	-27.9	27.9	55.8	-4.27	-14.9	5.24	15.2	-1.24	30.1	17.4	-17.4	34.8	7.89	-7.89	15.8
Fe	0.657	-7.87	10.7	-6.03	3.21	18.6	-26.0	26.0	52.0	-23.3	-16.8	-1.1	31	10.2	47.8	47.7	-47.7	95.4	-38.6	38.6	77.2
Ca	0.818	-4.42	7.41	-3.38	0.388	11.8	-6.99	6.99	14.0	9.01	-6.15	2.75	-22.3	16.7	39.0	32.4	-32.4	64.8	12.4	-12.4	24.8
Al	0.451	-5.76	23.0	-11.9	-5.33	34.9	-10.6	10.6	21.2	12.0	-16.4	-34.6	40.3	-1.31	74.9	7.55	-7.55	15.1	13.3	-13.3	26.6
Mg	0.529	-4.15	2.62	-8.98	10.5	19.5	-0.388	0.388	0.78	3.18	3.56	-15.7	-21.3	30.2	51.5	34.1	-34.1	68.2	3.27	-3.27	6.54
K	0.396	-2.98	4.18	-0.477	-0.722	7.16	-3.79	3.79	7.58	-3.44	0.497	-14.3	13.1	4.06	27.4	-9.68	9.68	19.4	-1.35	1.35	2.70
Cl	0.193	5.83	-12.3	9.83	-3.33	22.1	-5.71	5.71	11.4	13.0	-5.44	-9.11	4.21	-2.63	13.3	-2.15	2.15	4.30	3.62	-3.62	7.24
Ti	0.820	-6.28	10.6	-3.49	-0.871	16.9	-17.9	17.9	35.8	2.63	1.71	-6.17	5.34	-3.51	11.5	26.6	-26.6	53.2	-3.00	3.00	6.00
Ba	0.702	14.8	16.3	-21.5	-9.69	37.8	-0.523	0.523	1.05	-0.64	-18.4	30.2	15.5	-26.6	56.8	41.6	-41.6	83.2	6.26	-6.26	12.5
Na	0.366	10.0	10.8	-13.2	-7.69	24.0	-14.5	14.5	29.0	-2.68	-41.5	-34.6	14.9	63.9	105	15.8	-15.8	31.6	12.3	-12.3	24.6
Zn	0.412	0.918	11.0	-10.9	-1.06	21.9	-22.9	22.9	45.8	-11.0	33.7	-29.5	-23.9	30.7	63.2	22.9	-22.9	45.8	-2.73	2.73	5.46
Cu	0.655	-2.45	11.9	-14.6	5.15	26.5	-5.62	5.62	11.2	36.9	-15.2	-29	11.8	-4.51	40.8	60.3	-60.3	121	-17.6	17.6	35.2
Mn	0.655	-8.86	18.5	-6.38	-3.27	27.4	-78.3	78.3	157	-21.5	-10.8	40.6	-65.1	56.8	122	35.6	-35.6	71.2	-39.4	39.4	78.8
Br	0.262	17.5	-15.2	18.3	-20.6	38.9	-8.00	8.00	16.0	24.9	-25.1	8.59	-17.6	9.19	34.3	-4.86	4.86	9.72	-14.6	14.6	29.2
Ni	0.448	-20.6	1.16	-14.4	33.8	54.4	-113	113	226	-10.9	-23.9	-93.3	-44.3	172	265	140	-140	280	-77.0	77.0	154
Pb	0.762	5.26	10.3	-20.1	4.49	30.4	-51.5	51.5	103	13.8	-31.1	-10.8	-4.81	32.9	64.0	45.5	-45.5	91.0	-7.35	7.35	14.7
Cr	0.464	-3.04	-57.7	18.6	42.1	99.8	-331	331	662	-137	117	-195	-26.1	241	436	241	-241	482	-258	258	516
Cd	0.679	-28.0	32.8	-19.2	14.4	60.8	-11.3	11.3	22.6	10.7	-15.2	4.88	-14	13.6	28.8	23	-23	46.0	8.38	-8.38	16.8

[a] R: Multiple correlation coefficient adjusted for the degrees of freedom; [b] SP: Screw press; BP: Belt press; RP: Rotary press; DP: Multi disk; CE: Centrifuge;

[c] Coagulant, 1: polymer only; 2: polymer+ polyferric sulfate or polyaluminum chloride

* Category score, unit: %

* Black cells: p < 0.01

Table 2.7 Correlation coefficients between elements.

	C	O	H	N	P	Si	Fe	Ca	Al	S	Mg	K	Cl	Ti	Ba	Na	Zn	Cu	Mn	Br	Ni	Pb	Cr	Cd
C	1																							
O	0.746	1																						
H	0.873	0.621	1																					
N	0.260	-0.232	0.372	1																				
P	-0.707	-0.690	-0.557	0.189	1																			
Si	-0.636	-0.531	-0.672	-0.410	0.219	1																		
Fe	-0.783	-0.531	-0.692	-0.240	0.659	0.317	1																	
Ca	-0.480	-0.563	-0.422	-0.071	0.552	0.488	0.194	1																
Al	-0.454	-0.451	-0.415	-0.035	0.243	0.637	0.022	0.278	1															
S	-0.675	-0.700	-0.540	0.182	0.647	0.196	0.623	0.403	0.098	1														
Mg	-0.414	-0.445	-0.330	0.145	0.757	0.149	0.334	0.626	-0.007	0.443	1													
K	0.072	-0.023	0.051	0.262	-0.055	0.058	-0.179	-0.165	0.117	-0.164	-0.003	1												
Cl	0.068	0.087	0.065	-0.055	-0.110	0.013	-0.088	0.061	-0.005	-0.030	-0.121	-0.042	1											
Ti	-0.722	-0.711	-0.679	-0.181	0.521	0.714	0.539	0.665	0.390	0.609	0.349	-0.082	0.047	1										
Ba	-0.228	-0.388	-0.257	0.119	0.224	0.282	0.144	0.463	0.126	0.354	0.162	-0.289	0.020	0.437	1									
Na	-0.151	-0.314	-0.198	0.214	0.196	0.095	0.109	0.248	0.037	0.433	0.392	0.132	-0.038	0.186	0.126	1								
Zn	-0.362	-0.280	-0.385	-0.281	0.212	0.303	0.282	0.312	0.204	0.251	0.063	-0.060	-0.111	0.430	0.102	0.093	1							
Cu	-0.537	-0.606	-0.422	0.061	0.559	0.302	0.425	0.512	0.220	0.498	0.300	-0.085	-0.003	0.621	0.392	0.103	0.416	1						
Mn	-0.412	-0.251	-0.456	-0.349	0.218	0.404	0.360	0.219	0.096	0.103	0.152	0.066	-0.120	0.284	-0.060	-0.001	0.412	0.118	1					
Br	0.050	0.007	0.017	-0.059	-0.058	-0.039	0.056	0.008	-0.095	-0.093	-0.083	-0.009	0.406	0.021	-0.078	-0.083	-0.068	0.079	0.050	1				
Ni	-0.417	-0.371	-0.382	-0.125	0.322	0.305	0.366	0.306	0.190	0.324	0.122	0.030	-0.077	0.473	0.026	0.177	0.683	0.499	0.367	-0.083	1			
Pb	-0.682	-0.571	-0.715	-0.373	0.395	0.639	0.541	0.443	0.340	0.455	0.244	-0.009	-0.061	0.676	0.207	0.271	0.626	0.517	0.576	-0.048	0.730	1		
Cr	-0.364	-0.292	-0.368	-0.186	0.232	0.276	0.379	0.212	0.173	0.231	-0.010	-0.036	-0.097	0.503	-0.003	0.081	0.685	0.370	0.365	-0.019	0.842	0.630	1	
Cd	-0.405	-0.406	-0.352	-0.137	0.289	0.469	0.097	0.540	0.346	0.259	0.267	-0.002	-0.077	0.429	0.321	0.169	0.331	0.310	0.415	-0.138	0.321	0.521	0.210	1

*Black cells: correlation coefficient > 0.7; gray: correlation coefficient = 0.7–0.5; white cells: correlation coefficient < 0.5

*Black and gray cells: $p < 0.01$

2.3.4 Van Krevelen diagram of sewage sludge

The results from this study were plotted in a Van Krevelen diagram using C, H, and O values (Figure 2.3). The horizontal axis represents the O/C atomic ratio, while the vertical axis represents the H/C atomic ratio, which is relevant to the chemical composition [33]. This method has been used for fossil fuel assessment and interpretation of matter distributions in environmental samples [34]. The blue line indicates dehydration, the green line shows decarboxylation, and the red line is demethanation (in the direction of the arrow). Observation of the sample's position shift in the diagram can be used to infer the extent of reaction of the sample and the main reaction process. In addition to the results, plots for a range of biomass and coal samples, with representative data obtained from the literature, are also shown for reference [35].

The O/C ratio was in the range of 0.4–0.7 and the H/C ratio was in the range of 1.5–2.5 (Figure 2.3a). Most of the digested sludge was concentrated in the upper left corner, shown in the enlarged image in Figure 2.3b. Compared with undigested sludge, which mainly underwent decarboxylation, the O/C ratio of the digested sludge was lower than that of the undigested sludge. This indicated that the C, H, and O concentrations of the sludge were reduced through the digestion process, which had the greatest impact on the organic matter content in the sludge as fuel, while the sewage collection system had a smaller effect. Compared with the positions of other biomass types in the diagram, the positions of digested and undigested sludge were similar to those associated with refuse-derived fuel (RDF).
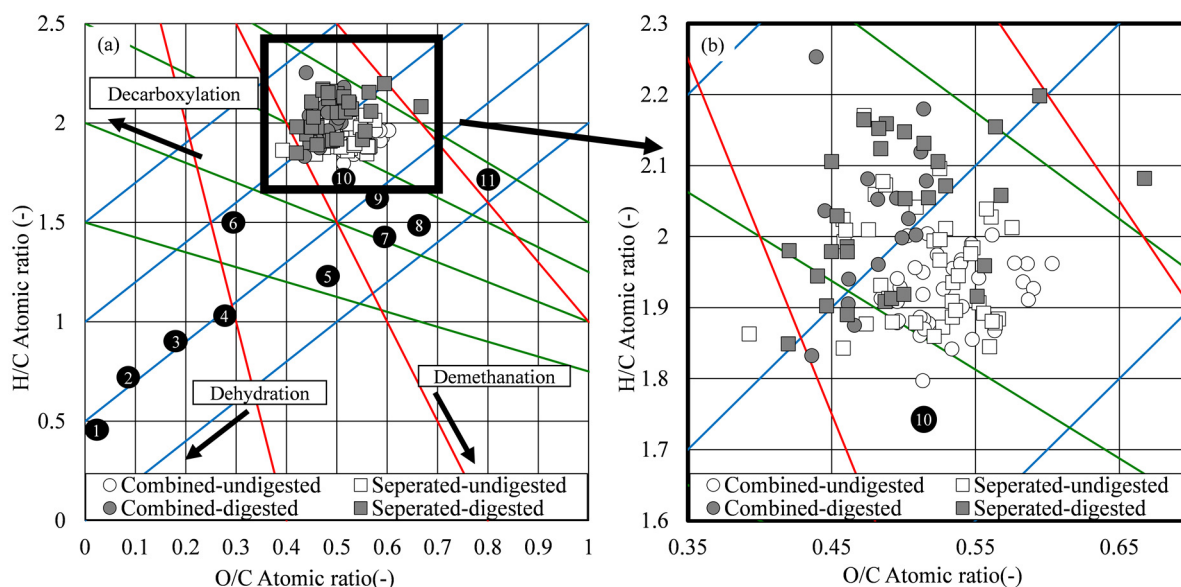

Figure 2.3 Van Krevelen diagram of sewage sludge and other reference materials: 1. anthracite, 2. bituminous coal, 3. sub-bituminous coal, 4. lignite, 5. peat, 6. manure, 7. woody biomass, 8. leaves and straw, 9. paper, 10. refuse-derived fuel, 11. algae. (a) Full size, (b) enlargement.

2.3.5 Principal component analysis and cluster analysis of the elemental composition

Principal component analysis (PCA) is a statistical method that converts the observations of a set of possibly related variables into a set of linearly uncorrelated variables, called principal components, and then uses only the first few principal components to reduce the dimensionality of the transformed data. Cluster analysis can then be used to group data by similarity. PCA and cluster analysis were conducted with the PAST software [36]. The PCA of the dewatered sludge elemental compositions for the 32 WWTPs is shown in Figure 2.4a for WWTPs and Figure 2.4b for elements. The two main principal components (PC1 and PC2) accounted for 76.8% and 13.9%, respectively, in total accounting for 90.7% of the data. In Figure 2.4a, PC1 showed large C and O contents, as well as N and H contents, and this axis was considered to represent organic matter. In PC2, C, N, Ca, Al, H, P, Mg, and S showed a positive influence, and this axis was presumed to represent household and domestic wastewater.

In Figure 2.4b, PC1 showed a high degree of influence from undigested processes, because many WWTPs without (with) digestion were found in the positive (negative) direction of PC1. The judgment criteria for PC2 was less clear; however, many WWTPs with a separated sewage collection system were found in the positive direction, and those with a combined system in the negative direction, indicating some degree of influence from separated systems. The results were reasonably consistent with those from Hayashi's quantification method I analysis.

The cluster analysis of the dewatered sludge element composition results is shown in Figure 2.5a for elements and in Figure 2.5b for WWTPs. In Figure 2.4a, the elements were clustered into four groups: C and O are located at the right of the graph; N and H are located at the right of the graph; P, Si, and Fe are located at the left of the graph; and the other elements are located around the origin. This spread has the same trend as the clusters in Figure 2.5a. In Figure 2.5b, the 32 WWTPs were clustered into three main groups: group 1 included 10 WWTPs, all of which used a digestion process (WWTP numbers 2, 3, 6, 7, 13, 14, 15, 19, 24, and 27); group 2 included 13 WWTPs, of which 2 used a digestion process and used no digestion process (4, 5, 8, 9, 16, 17, 18, 22, 23, 26, 28, 29, and 32); and group 3 included 9 WWTPs, all of which used no digestion process (1, 10, 11, 12, 20, 21, 25, 30, and 31). In group 2, two-thirds of the WWTPs ad a separated collection system, whereas group 3 contained 50% separated and 50% combined collection systems. From the results of the PCA and cluster analysis, the sludge digestion process has a greater effect on the composition of dewatered sludge than the sewage collection system and coagulants for dewatering.

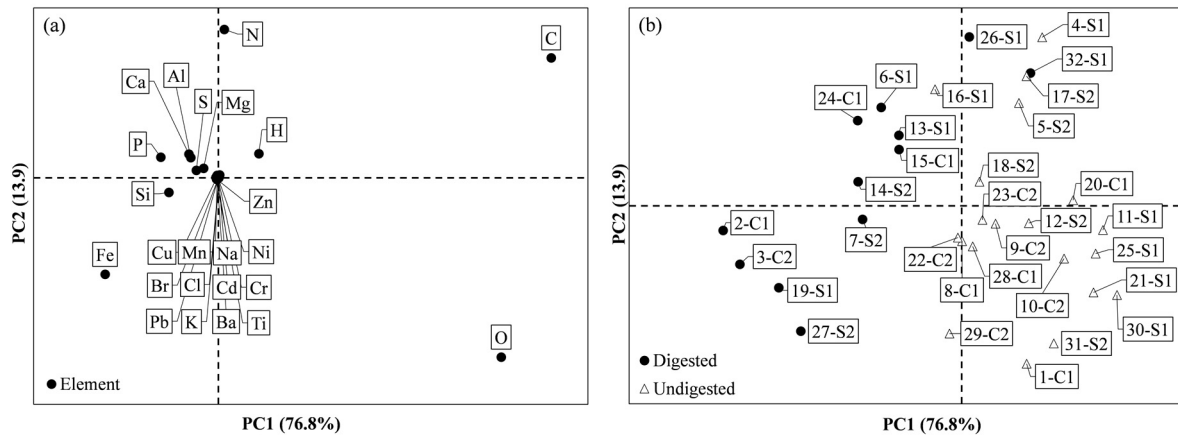Figure 2.4 Principal component analysis of the elemental compositions of dewatered sludge (a) elements and (b) 32 wastewater treatment plants. S: separated, C: combined, 1: polymer, 2: polymer+ polyferric sulfate or polyaluminum chloride.
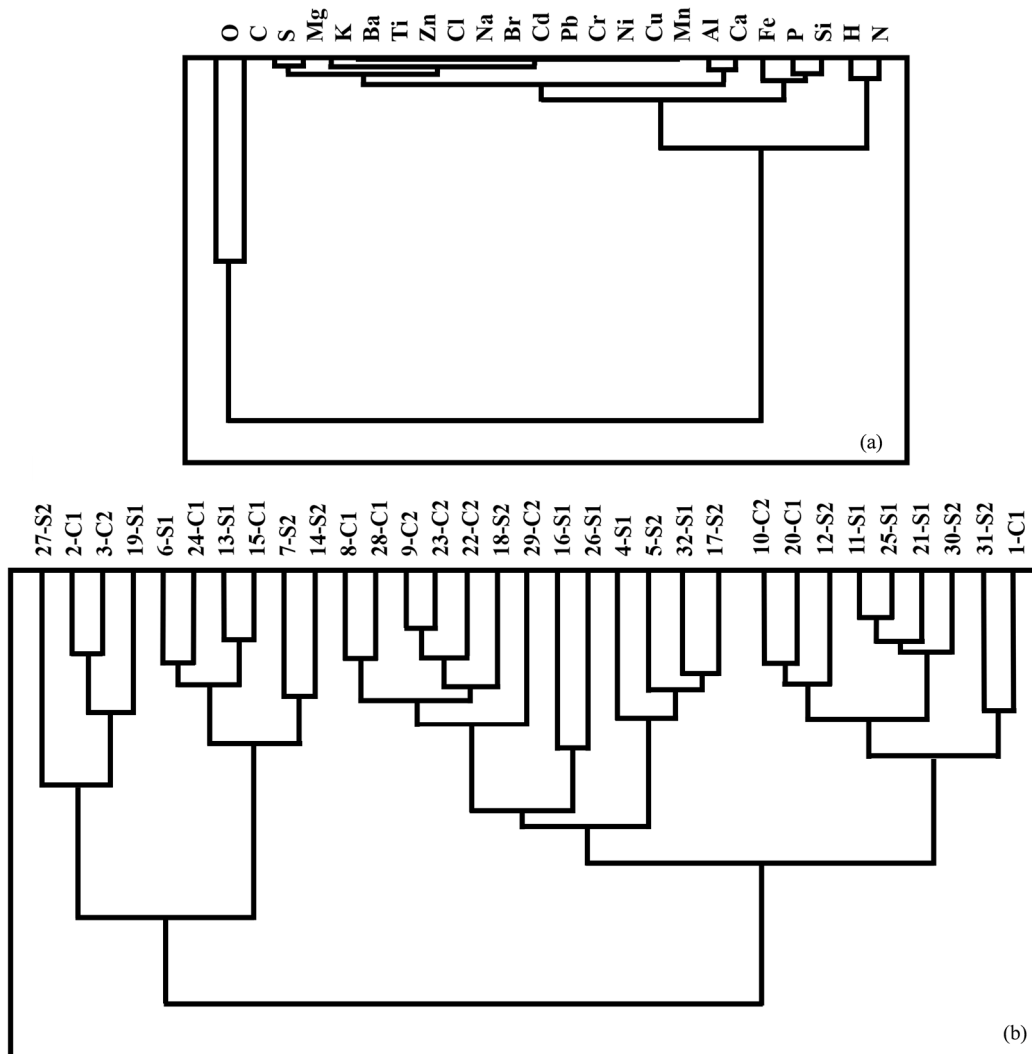

Figure 2.5 Cluster analysis of (a) elements and (b) wastewater treatment plants. S: separated, C: combined, 1: polymer, 2: polymer + polyferric sulfate or polyaluminum chloride.

2.3.6 Standard sewage sludge composition and potential for practical application

According to the results described above, the sewage collection system and digestion process used in the WWTPs were significant factors influencing sludge composition. Thus, the standard composition of dewatered sludge can be classified according to the four categories summarized in Table 2.8, which provides a useful reference for sludge compositions from WWTPs.

The heavy metal content of digested sludge is higher than that of undigested sludge, and that of sewage from combined collection systems is higher than that from separate systems. The Cr, Cu, Pb content of the digested sludge is about twice that of the undigested sludge, regardless of whether it is combined or separated. The Ni and Zn contents of digested and undigested sludge in the separated system (Ni: 24.6 and 29.9 mg/kg, Zn: 537 and 561 mg/kg, respectively) do not differ significantly. However, in the combined system, the Ni and Zn contents of digested sludge are much higher than those of undigested sludge (Ni: 24.8 and 159 mg/kg, Zn: 600 and 1,259 mg/kg, respectively).

As mentioned in 2.3.1, many countries have heavy metal standards for sludge recycling in agriculture. Undigested sludge conforms to the Japanese regulations regardless of whether a separated or combined sewage collection system is used. The Cu content of digested sludge in the combined and separated system is 577 and 517 mg/kg, respectively, which exceed the standard of 300 mg/kg. The Zn content in the combined digested sludge, of 1,295 mg/kg also exceeds the standard of 900 mg/kg. In summary, in terms of the perspective of heavy metal content, regardless of the sewage collection system, undigested sludge has great potential for agricultural recycling. The Cu and Zn contents of digested sludge (especially combined digested sludge) need to be addressed before it can be considered for use.

The possibility of recycling sewage sludge in the cement industry is also being studied. In addition to promoting the recycling of waste sludge, an advantage of sludge recycling is that the combustion of fuel in a rotary cement furnace is also a "non-waste process" because the ash from combustion can be used as clinker [12]. Rezaee et al. [37] proposed that the silicon, aluminum, and iron oxide content of sewage sludge makes it a suitable replacement for some of the raw materials used during clinker production. Mokrzycki et al. [12] reported the following criteria for the use of alternative fuels in the cement industry: heating value over 14.0 MJ/kg-DS, Cl content less than 0.2%, S content less than 2.5%, polychlorinated biphenyls (PCB) content less than 50 ppm, and heavy metals content less than 2,500 ppm [including mercury (Hg) content less than 10 ppm, and total Cd, thallium (Tl) and Hg content less than 100 ppm]. The criteria for use of alternative fuels in the cement industry are summarized in Table 2.9. The heating values of the four categories of sludge are in the range of 17,800~18,270 kJ/kg-DS, which meets the requirement of being greater than 14,000 kJ/kg-DS. The S, Cl, and heavy metal (Zn, Cu, Ni, Pb, Cr, Cd) contents also conform to the criteria. This indicates that, regardless of the sewage collection system and digestion status, sludge could potentially be used as an alternative fuel in the cement industry. In addition, EN450-1 stipulates that the P_2O_5 of coal ash and co-incineration ashes in concrete should not exceed 5.5wt%. Although the use of sludge is

not covered by the standard EN450-1, high P content may limit the application of sewage sludge incineration ash as a cementitious material [38].

Table 2.8 Properties and elemental compositions of sewage sludge by category.

Sewage collection	Combined		Separate	
Digestion process	Undigested	Digested	Undigested	Digested
HHV(kJ/kg-DS)	17,900	18,100	18,300	18,200
LHVd [a] (kJ/kg-DS)	16,400	16,700	16,800	16,700
LHVw [b] (kJ/kg)	1,060	1,580	1,500	1,380
M (%)[c]	77.6	80.9	80.0	81.5
V (%)[d]	81.6	70.0	85.5	75.6
C	407,000	350,000	424,000	374,000
O	286,000	218,000	287,000	242,000
H	65,200	59,000	69,300	63,200
N	47,600	54,200	63,100	60,700
P	22,600	34,800	24,500	33,700
Si	24,400	33,200	15,000	21,400
Fe	20,200	37,300	13,200	27,900
Ca	11,200	19,500	9,200	17,200
Al	11,500	16,100	11,300	12,400
S	10,100	18,800	10,500	16,500
Mg	4,840	9,390	4,880	8,310
K	3,820	3,620	3,840	2,950
Cl	1,280	1,110	1,100	1,160
Ti	987	1,540	681	1,160
Ba	602	1,190	668	1,190
Na	658	1,620	863	748
Zn	600	1,300	537	561
Cu	229	577	247	517
Mn	573	692	178	326
Br	86.3	53.0	65.9	71.6
Ni	24.8	159	24.6	29.9
Pb	21.4	44.2	8.55	19.5
Cr	51.9	144	10.6	25.8
Cd	0.87	1.46	0.753	1.11
Number	35	16	40	29

*Unit: mg/kg

[a] LHVd: LHV of dried sample

[b] LHVw: LHV of dewatered sample

[c] M: moisture content

[d] V: volatile matter

Table 2.9 Criteria for the use of alternative fuels in the cement industry [12].

Unit (DS)		Criteria [12]	Combined		Separated	
			Undigested	Digested	Undigested	Digested
HHV	kJ/kg	14,000	17,900	18,100	18,300	18,200
S	%	2.5	1.10	2.18	1.12	1.86
Cl	%	0.2	0.14	0.13	0.12	0.13
Cd	mg/kg	100	0.87	1.46	0.75	1.11
Heavy metal [a]	mg/kg	2,500	928	2,220	828	1160

[a] Heavy metal including Zn, Cu, Ni, Pb, Cr and Cd.

2.4 Conclusion

This study demonstrated that the elemental composition of sludge in Japan is similar to that in other countries. The contents of the elements in sludge ranged widely, with large deviations. The Cu concentration of dewatered sludge in Japan exceeds the regulatory limits for agricultural use, which warrants caution. The results revealed that the sewage collection system, digestion process, and coagulant used for dewatering affected the performance of dewatered sludge and the elemental concentrations. However, seasonal effects and the dewatering device had little influence on elemental concentrations. The digestion process had the strongest association with the elemental composition of the dewatered sludge. In particular, the digestion process has a major impact on organic matter content. The dewatered sludge from the 32 WWTPs could be divided into three major categories. The elemental correlations were likely influenced by natural sources and industrial wastewater rather than domestic wastewater. In addition, the compositions of sludge from WWTPs both with and without digestion were similar to RDF and almost comply with the biomass solid fuel -15 (BSF-15, which the heating value higher than 15 MJ/kg) of Japanese Industrial Standards. Based on the sewage collection system and digestion status, standard compositions of sludge in these four categories were proposed. The heavy metal content of the sludge from the combined system is higher than that of the separate systems and is greater in digested sludge than in undigested sludge. Undigested sludge has greater potential than digested sludge for agricultural recycling. According to the criteria of Lafarge Cement Polska, these four types of sludge could be used as an alternative fuel for the cement industry. However, due to the high P content, there are still doubts as a raw material for cement.

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Chapter 3 Co-incineration effect of sewage sludge and municipal solid waste on the behavior of heavy metals

3.1 Introduction

Sewage sludge and Municipal solid waste (MSW) contain hazardous heavy metals and other useful elements. Generally, it has been known from past studies that chlorine (Cl) easily combines with heavy metals to form chlorides and migrate to gases at high temperatures, which increases the volatilization of heavy metals [1]. Liu et al. [2] investigated the volatilization of heavy metals including nickel (Ni), lead (Pb), cadmium (Cd), chromium (Cr), copper (Cu), zinc (Zn), manganese (Mn) by adding different ratios of Cl and phosphorus (P), to clarify the effects of Cl and P on the volatilization of heavy metals during the mono-incineration of sulfur (S)-rich textile dyeing sludge. They indicated that at a certain concentration of P, its effect inhibits the volatilization of heavy metals bonded to Cl. In addition to Cl and P, the S content during sludge or MSW mono-incineration also affects the chemical states of heavy metals [3][4]. Tang et al. [5] reported that adding hydroxyapatite (HAP) during the incineration of tannery sludge can reduce the volatilization of heavy metals, and much more thermostable phosphate minerals such as $CrPO_4$, $Cu_2(PO_3)_4$-$9H_2O$, and $Cu_3(PO_4)_2$ were found in bottom ash. Therefore, the presence of P plays a certain role in the stability of heavy metals during incineration.

Co-incineration of sewage sludge and MSW has been shown by some studies to be more effective than mono-incineration of each waste [6][7]. However, these studies were focused on the calculation of greenhouse gas emissions and cost as a lifecycle inventory analysis. The interaction effect on co-incineration of sewage sludge and MSW has not been clarified, especially heavy metals have a high rate of transfer to exhaust gas during high-temperature incineration [8]. The composition of sewage sludge and MSW causes different incineration characteristics [9][10], and which on co-incineration also needs to be confirmed. In addition, during the co-incineration of these two different wastes, if a compound with a higher boiling point is generated, it will tend to remain in the bottom ash; if a compound with a lower boiling point is generated, the residual ratio in the ash will become lower.

Therefore, co-incineration may have an interaction effect that cannot be obtained by mono-incineration and the interaction between the co-incineration of sewage sludge and MSW should be clarified. Chen et al. [11] used model sewage sludge and MSW for co-incineration in China. The results indicated that that as the blend of sewage sludge increases, the mild acid-soluble fraction of heavy metals such as arsenic (As), Cu, Zn, and Cr in the bottom ash decreases, and the residual fraction increase. This means that co-incineration can transform the fraction of heavy metals into a more stable residual fraction. But As has the opposite trend. The high temperature also has a stabilizing effect on the form of heavy metals in the bottom ash. In addition, another study by Chen et al. [12] also explored the volatilization characteristics of heavy metals including As, Cr, Cu, Ni, Pb, and Zn during co-incineration with or without calcium-based sorbents. The results indicated that calcium-based sorbent decreased the

volatilization of As, Cr, Cu, Ni, and Zn, but enhanced that of Pb.

Although Chen et al. discussed the impact of co-incineration of sewage sludge and MSW on heavy metals in China, the used MSW included 37.7% of food, 28.5% of paper and textiles, and 25.5% of plastic, the composition, management, and treatment of MSW have different definitions and classifications in various countries. Figure 3.1 shows the composition of MSW in some research which includes the U.S, Canada [13], South Africa [14], Iran [15], UK [16], EU [17], and Japan [18]. The variability of MSW in different regions is quite large. In addition, according to the data collected by The World Bank, the composition of MSW differs depending on national income level, and the lower the income, the greater the proportion of organic matter (Figure 3.2). High-income countries are expected to have no significant change in MSW in 2025, which because of the implementation of the reduce, reuse, recycle (3R) policy [19].

It's known from the above literature review that high P content helps stabilize heavy metals during incineration. But the previous research [4][5] were based on the addition of know substances such as $NH_4H_2PO_4$ or HAP. At present, there is no research on the influence of P contained in sludge on heavy metals during co-incineration with MSW. From the results in Chapter 2, it was found that the P content in sewage sludge is as high as about 2%, the interaction effect of P and heavy metals during co-incineration is worth looking forward to. This study is to investigate the co-incineration behavior change compared with mono-incineration by thermogravimetric analysis, and the behavior of Cu, Pb, Cd, and P on sewage sludge and MSW co-incineration based on the different waste compositions.
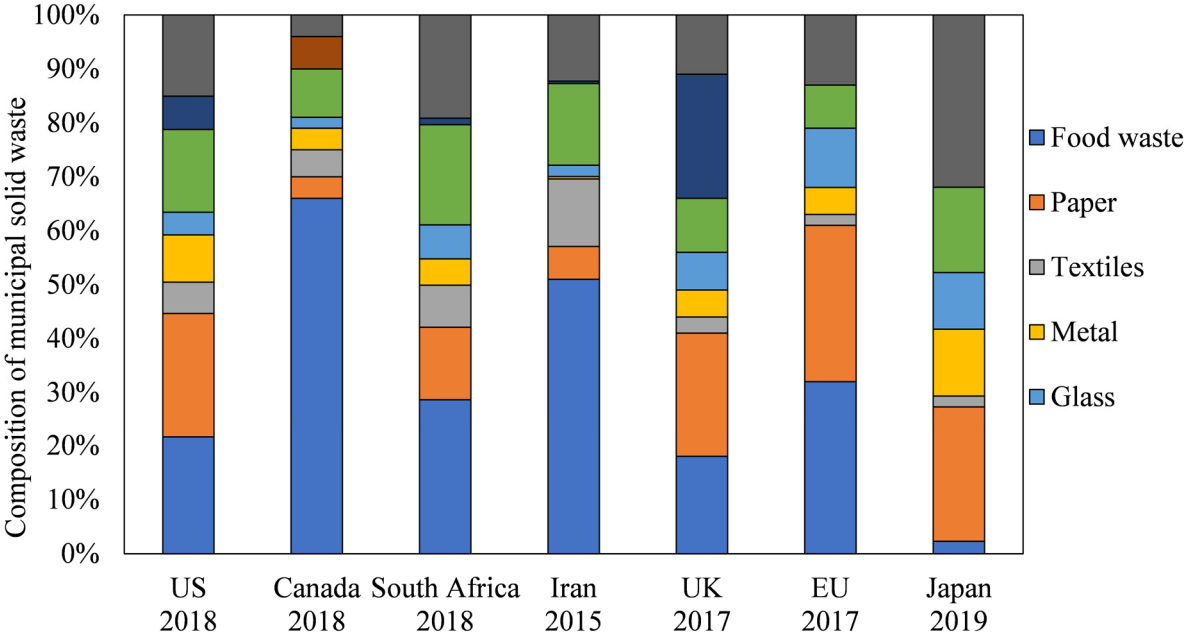

Figure 3.1 Composition of MSW from several research [13][14][15][16][17][18].
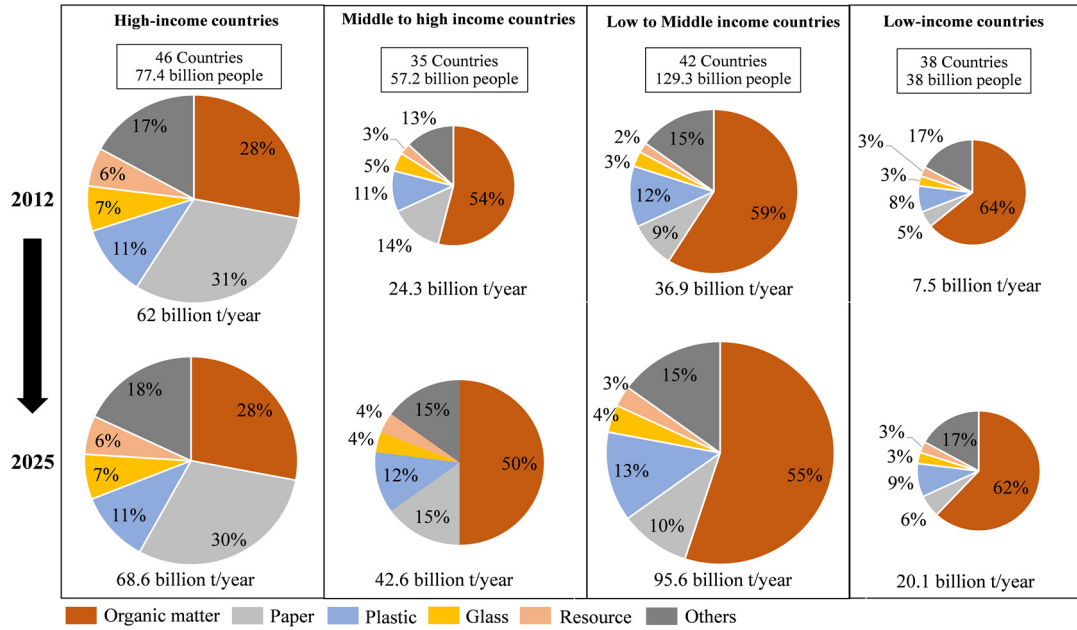
Figure 3.2 MSW composition and its prediction in different income level countries [19].

3.2 Materials and methods

3.2.1 Materials

Sewage sludge was collected after the sludge dewatering process in a wastewater treatment plant in Japan. The sewage collection system is separated collection (separated rainwater from sewers), and the sewage biological treatment method is Anaerobic-Anoxic-Oxic process. The undigested sludge produced is only added with diluent and polymer and dewatered by belt filtration. MSW samples (Refuse-derived fuel: RDF pellets) were obtained from a plant located in west Japan which were produced from MSW through a series of shredding and sorting processes followed by sanitary treatment under steam pressure, after that, the MSW was dried and formed into 15 mm diameter pellets. However, in this laboratory experiment, RDF pellets were powder-milled and sieved under 2 mm for homogenization of the sample. Both sewage sludge and MSW were dried at 105°C for 24h and crushed into powder with a particle size of <75μm. In this study, the dried sludge was blended into the treated MSW in different proportions on a dry basis weight, which included 10%, 20%, 50% of sewage sludge blending.

3.2.2 TG-DTA analysis

Thermogravimetric and differential thermal analysis (TG-DTA) was carried out using a differential thermogravimetric analyzer (Thermo Plus EVO 2, TG 8120; Rigaku). 5 mg-DS of the original and mixed samples were heated from ambient temperature to 900°C at a rate of 10°C/min under an air atmosphere. All analyses were conducted in duplicate or triplicate and the average was taken.

3.2.3 Co-incineration experiment

A tubular furnace (Φ=45mm, L=1000mm) was used in the co-incineration experiment (Figure 3.3). Highly purified air (O_2 21%, N_2 79%; Air-Zero-A, Sumitomo Seika, Osaka, Japan) flowed into the furnace at 200 mL/min. A sample (5g) was placed on a quartz boat (10cm, 30mL) and set in the center of the tubular furnace. The vaporized fraction of the heavy metals was collected in glass wool at the end of the quartz tube and in two bottles of collection liquid (5N HNO_3) behind the quartz tube. All samples were combusted at the target temperature (900°C) for 2 hours which included 1 hour of the heating process (heating rate:15°C/min), and the excess air ratio is 1.0 - 1.1. The ash left on the quartz boat was defined as bottom ash, and the heavy metals and phosphorus remaining in the bottom ash were analyzed by ICP-AES after acid digestion. All analyses were conducted in duplicate or triplicate and the average was taken.
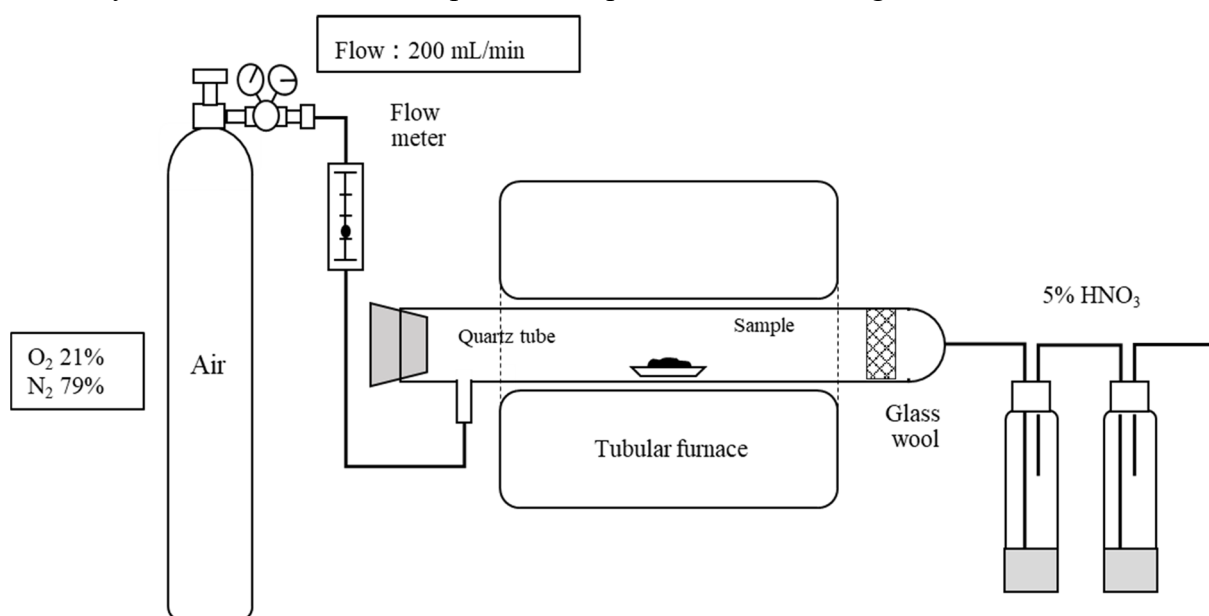
Figure 3.3 Schematic of the tubular furnace in the co-incineration experiment.

3.2.4 Analytical methods

3.2.4.1 Proximate analysis and ultimate analysis

Proximate analysis of sewage sludge and MSW was determined according to the Japan Industrial Standard [20][21]; ultimate analysis of carbon (C), hydrogen (H), nitrogen (N), and sulfur (S) was conducted by a CHN analyzer (JM-10; J-Science Lab Co. Ltd., Kyoto, Japan) [22] and an X-ray fluorescence spectrometer (XRF; XRF-1800; Shimadzu Co. Ltd., Kyoto, Japan). However, the XRF analyzer could not measure the O percentage accurately, so the percentage of O was calculated using Eq. (3.1).

$$O = V - C - H - N - S \qquad (3.1)$$

Where the unit is % (dry base) of each element.

3.2.4.2 Heavy metals and P analysis

Heavy metals including Cu, Pb, Cd, and P were analyzed after acid digestion. A dry sample weighing 0.5 g was placed in a 100-mL Erlenmeyer flask. HNO_3 (69%, SP; Nacalai Tesque, Inc., Kyoto, Japan) and HCl (35%, SP; Nacalai Tesque, Inc., Kyoto, Japan) were added in a volume ratio of 1:1. The mixture was then heated on a hot plate to almost dryness and HNO_3 and HCl were added in the same ratio. The above procedures were repeated until the liquid became light yellow and the precipitate was grayish. Metal concentrations were analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES) (ICPS-8000; Shimadzu Co. Ltd., Kyoto, Japan). All analyses were conducted in duplicate or triplicate and the average was taken.

3.2.4.3 Leaching test

The Japanese leaching test No. 13 (JLT-13) was conducted to determine the leachability of heavy metals in bottom ash. Ash sample and solvent (distilled water) were mixed in a ratio of 1:10 (g/mL), and continuous horizontal shaking for 6 hours with the velocity of 200 rpm (round per minute) at room temperature. After shaking, the leachate was filtered with a 1 μm pore size glass fiber filter (Advantec, Toyo Roshi Kaisha, Ltd., Japan). Afterward, the leachate was analyzed to determine heavy metals (Cu, Pb, and Cd) by inductively coupled plasma mass spectrometry (ICP-MS, Xseries2, Thermo Fisher Scientific, USA).

3.2.5 X-ray adsorption fine structure measurement (XAFS)

The chemical states of Cu in the raw sample and ash sample after co-incineration were detected by in situ Cu K-edge XAFS (BL-12C in Photon Factory, Tsukuba, Japan). Cu K-edge (8763 - 9985 eV) spectra of the samples were measured. The merged spectra were normalized by software Athena (ver. 0.9.25), and XAFS spectra of Cu in Cu-foil, Cu (311) crystals, CuO, Cu_2O, CuS, Cu_2S, CuCl, $CuSO_4$, $CuSO_4$-H_2O, $CuCl_2$, $CuCl_2$-$2H_2O$, $CuCO_3$, $CuFe_2O_4$, $Cu(OH)_2$, $Cu_3(PO_4)_2$, $CuBF_4$-$6H_2O$-Trans, Cu-II diethyldithiocarbamate, $Cu(OCOCH_3)_2$, CuBr, $CuBr_2$, $Cu[(H_2O)(Me_3TPA)](ClO_4)_2$, Atacamite, Dioptase, and Turquoise were measured as reference spectra, for comparing shapes and identifying the species.

3.3 Results and discussion

3.3.1 Characteristics of sewage sludge and MSW

Table 3.1 shows the sludge and MSW characteristics used in this study and Chen's study [12]. In this study, MSW has a higher concentration of fixed carbon and a lower content of volatile matter compare to sludge. The concentration of N, P, S in sludge is higher than that in MSW, and the Cl is higher in MSW. The concentration of Cu in sludge and MSW are approximately the same. As for Pb, Cd in MSW is greater than that in sludge. The higher heating value (HHV) of this study in sludge and MSW is higher than which in Chen's study, and the volatile content

of the sludge in this study is 88.9%, which is significantly different from the 28.1% of Chen's study. In addition, the concentration of Cu and Pb in this study is higher in MSW than in sludge, but which is the opposite in Chen's study. As mentioned above, P also affects the migration of heavy metals. However, Chen's study did not mention this part.

Table 3.1 Composition of sewage sludge and MSW in this study and Chen's study [12].

Parameter	Unit	This study (Japan)		Chen's study (China)	
		Sludge	MSW	Sludge	MSW
HHV	MJ/kg	17.7	19.6	5.61	15.9
Ash	%	10.0	8.6	68.1	6.42
Volatile matter	%	88.9	81.1	28.1	79.3
Fixed carbon	%	1.1	10.3	3.82	14.3
C	%	44.2	45.0	13.3	38.8
H	%	5.60	6.49	2.04	5.52
N	%	6.70	1.06	2.13	2.25
O	%	31.7	28.4	14.1	39.2
Cl	%	0.068	0.757	0.18	7.79
S	%	0.684	0.137	0.15	-
P	mg/kg	19900 ± 3500	973 ± 23.6	-	-
Cu	mg/kg	249 ± 4.77	228 ± 3.25	127	31.9
Pb	mg/kg	8.04 ± 5.1	17.2 ± 4.08	41.9	3.75
Cd	mg/kg	0.34 ± 0.54	0.822 ± 0.1	-	-

3.3.2 TG-DTA analysis

Figure 3.4a and Figure 3.4b show the DTG and DTA curves of the samples, respectively. The theoretical value (black line) indicates the value calculated based on the ratio of MSW and sludge blending. The calculation shows as Eq. (3.2) and (3.3).

$$DTG_{th} = DTG_{MSW} \times \left(1 - B(\%)\right) + DTG_{sludge} \times B(\%) \qquad (3.2)$$
$$DTA_{th} = DTA_{MSW} \times \left(1 - B(\%)\right) + DTA_{sludge} \times B(\%) \qquad (3.3)$$

Where DTG_{th} (%/min) and DTA_{th} (μV) are the theoretical value, DTG_{MSW}/DTA_{MSW} and $DTG_{sludge}/DTA_{sludge}$ are the DTG and DTA result (experimental value) of MSW and sludge, B is the blend ratio of sludge.

By comparing with the experimental value (color line), the interaction effect that occurred in co-incineration can be fined. In Figure 3.4a and Figure 3.4b, there are two obvious weight loss peaks of MSW in DTG, from 200°C to 380°C and from 400°C to 500°C, and the maximum weight loss rate is at 300°C and 460°C, respectively. The first peak represents the release of organic matter, weight loss of 300°C shows the combustion of organic matter. The second peak represents the oxidation and thermal decomposition of polymer and fixed carbon [23][24]. Furthermore, there are three exothermic peaks in DTA. The temperature of 200°C~380°C

belongs to the volatilization and incineration zone of organic matter, but the ignition point changes with various compositions of material. The peaks at 250°C and 320°C in DTA can be inferred that it may come from different organic sources. The volatile matter in the cellulosic matter is easier to decompose and combusted at a low temperature than the volatile matter in plastic [25]. Therefore, the peaks at 250°C and 320°C are likely to come from cellulosic and plastic.

DTG curve of the sludge only showed one obvious weight loss peak at 300°C. As the blend ratio of sludge increases, the first peak gradually increases, and the second peak gradually weakens. This result concurs with Table 3.1, compared to MSW, the combustion behavior of sludge is mostly caused by volatility, while the fixed carbon is less, and MSW is caused by both organic matter and fixed carbon. Compared to the theoretical value, the weight loss peak at 460°C during co-incineration is a little shifted to the high temperature with the blending ratio of sludge. At 300°C, the organic matter in the sludge volatilizes during co-incineration, and the oxygen in the air is consumed for the combustion of the organic matter. After that, oxygen arrives at the fixed carbon in the solid and combust. Therefore, if the proportion of sludge is high, it takes time to decompose the volatile organic matter, which delay the decomposition of polymers and fixed carbon at 460°C.
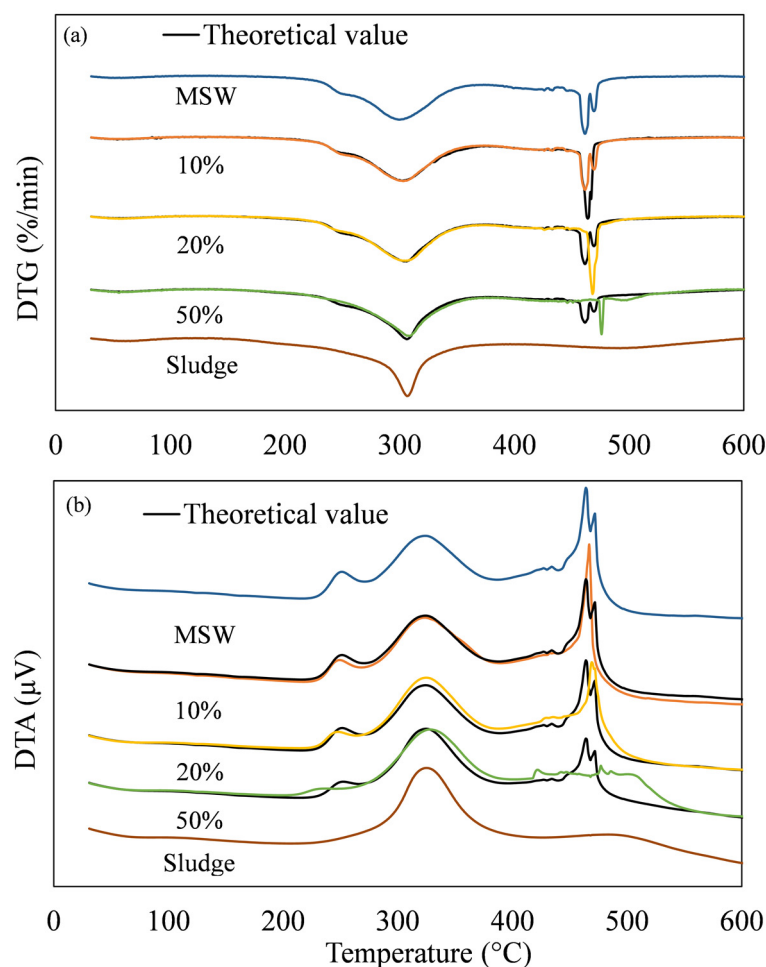

Figure 3.4 (a) DTG and (b) DTA curve of samples.

3.3.3 Migration characteristics of heavy metals during co-incineration

The residual ratio of heavy metals in bottom ash after incineration was calculated as Eq. (3.4)

$$RR = \frac{C_{ash} * A}{C_{raw}} \qquad (3.4)$$

Where *RR* (%) is the residual ratio in the ash, C_{ash} and C_{raw} are the concentration of heavy metal in ash/raw sample (mg/kg), A is the ash content (%) of each raw sample.

Table 3.2 shows the concentration of Cu, Cd, Pb, and P in the raw sample, ash sample after the co-incineration experiment. For comparison, the table also lists the ash sample which was expressed based on the same weight as the raw samples. Figure 3.5 shows the residual ratio of heavy metals under different blending ratios after incineration. The theoretical value (black dotted line) indicates the value calculated based on the ratio of sludge and MSW concentration blending. The calculation is shown as Eq. (3.5). By comparing with the experimental value (color line) of incinerating at 900°C, the effect of co-incineration on heavy metals can be fined.

$$C_{th} = C_{MSW} \times \left(1 - B(\%)\right) + C_{sludge} \times B(\%) \qquad (3.5)$$

Where C_{th} (%) is the concentration of theoretical value, C_{MSW} and C_{sludge} are the concentration in MSW and sludge, B is the blend ratio of sludge.

Cu concentration of sludge and MSW is similar in raw samples. In the case of MSW mono-incineration, the residual ratio was 18%, which was as high as 91% in the sludge mono-incineration. The co-incineration residual ratio was higher than the theoretical value, which indicates that the co-incineration has the effect of remaining Cu in the ash. The residual ratio of Cd was 5% in MSW and 44% in sludge. Compared with MSW, Cd in sludge tends to remain in the ash. However, as the proportion of sludge added increases, the residual ratio of Cd was lower than the theoretical value, which indicates that co-incineration increased the volatilization of Cd. Pb was almost volatilized, and the residual ratio was only 2% in MSW mono-incineration. However, the residual ratio in sludge mono-incineration was 65%. The residual ratio of co-incineration almost agrees with the theoretical value, which indicates that co-incineration did not seem to have much effect on the migration of Pb.

The element of Cu is considered to be a non-volatile metal, and Cd and Pb are volatile metals [26], which is the reason why the residual ratio of Cu in the mono-incineration of MSW and sludge is higher than that of Cd and Pb, the same results are also shown in the previous survey [3][27]. According to the literature, these three heavy metals are easily affected by Cl [28], and the degree of volatility is Cd > Pb > Cu [3]. It is quite easy to form a metal chloride with lower boiling points and evaporate. Liu et al. [2] also reported that Cl would increase the volatilization of Pb, but the presence of P would make Pb form residual fractions and stay in the bottom ash. The MSW used in this study has high Cl content (0.75%) and low P content (0.097%), while

the sludge has low Cl content (0.068%) and high P content (2%). It can be inferred that, possibly due to the influence of Cl, the residual ratio of these three heavy metals in sludge mono-incineration is higher than that in MSW mono-incineration. And due to the strong interaction between the particles in the metal phosphate, the presence of P has helped solidify trace heavy metals in sludge incineration [3], therefore, P may also play a role in solidifying heavy metals in this experiment. In the coagulation unit of sewage treatment, $Ca(OH)_2$ may be added to help adjust the alkalinity, which will cause the organic P in the wastewater to precipitate in the form of apatite, resulting in lower vaporization [29]. This may explain the phenomenon that P almost remains in the bottom ash.
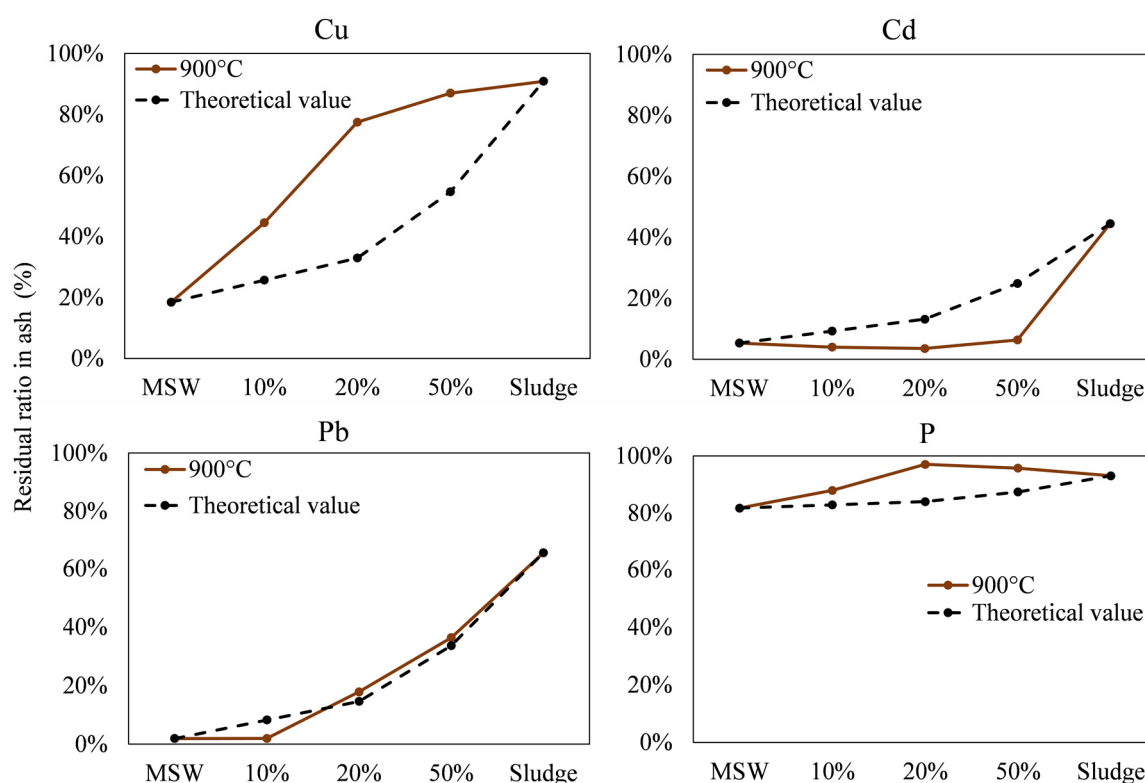
Figure 3.5 Residual ratio of heavy metals under different blending ratios after incineration.

Table 3.2 Concentration of Cu, Cd, Pb, and P in the raw samples and the ash samples.

Sample		Cu	Cd	Pb	P	Ash content
Raw sample (mg/kg)	MSW	228	0.82	17.2	973	
	10%	381	0.79	20.8	2890	
	20%	168	0.84	16.0	4710	
	50%	178	0.72	11.5	10300	
	Sludge	249	0.34	8.04	19900	
Ash sample (mg/kg-raw sample base)	MSW	42	0.044	0.32	795	8.63%
	10%	169	0.032	0.40	2550	8.93%
	20%	130	0.030	2.85	4570	9.16%
	50%	155	0.046	4.19	9900	9.81%
	Sludge	226	0.151	5.26	18500	9.86%

3.3.4 Leaching characteristics of heavy metals in incineration ash

Table 3.3 shows the results of the leaching test, the standard value refers to the criteria for landfill disposal of industrial waste in Japan [30]. The leaching concentration of Cu and Pb in MSW ash was higher than that of sludge and co-incineration ash, and the concentration of Cd was almost undetectable in the leachate. Overall, the leaching ability of Cu, Cd, and Pb were low, and all samples do not exceed the standard values of Cd and Pb. Although the concentration of Cu is not specifically specified, the result of the leaching test was quite low, which refers to having no impact on the landfill and environment.

Table 3.3 Heavy metals concentration in leachate.

Sample	Heavy metals concentration (mg/L)		
	Cu	Cd	Pb
Standard	-	0.09	0.3
MSW	0.13	<0.0001	0.0093
10%	<0.001	<0.0001	0.0003
20%	<0.001	<0.0001	0.0009
50%	0.0172	<0.0001	0.0051
Sludge	0.0341	<0.0001	0.0014

3.3.5 XAFS measurement

The X-ray Absorption fine structure (XAFS) spectra of raw samples, ash samples after incineration, and Cu standard substances (CuO, Cu_2O, $CuSO_4$, $CuSO_4$-H_2O, $CuCO_3$, $Cu_3(PO_4)_2$) are shown in Figure 3.6, and the spectra of other Cu standard substances could be found in the appendix (Figure A3).

Cu standard substances have obvious peaks in 8970–9010 eV. Compared with $CuSO_4$, $CuSO_4$-H_2O, $CuCO_3$, $Cu_3(PO_4)_2$, the peaks of CuO, Cu_2O are relatively flat, and have obvious small peaks at 8985 and 8980, respectively. The peak of CuO is about 8997 eV, and the other peaks are about 8994 eV. However, there are some slight differences in the peaks of other peaks at 8994 eV like $CuSO_4$, $CuSO_4$-H_2O are sharper, and $Cu_3(PO_4)_2$ is gentle. In addition, $CuSO_4$ and $CuSO_4$-H_2O have a small characteristic peak at 8988 eV. These differences can help us determine the chemical states of Cu in the samples.

It can be seen from the figure that the peaks of the raw sludge and MSW are at 8997 eV, and there is a small characteristic peak at 8985 eV in both sludge and MSW, which is consistent with the characteristics of CuO. Mono-incineration ash of sludge and MSW have obvious differences in 8975–8990 eV. The peak of sludge is steeper and the MSW is gentler, and the wave crest of sludge is more to the left than that of the MSW. The ash of MSW may be close to the CuO, and sludge may be similar to the $Cu_3(PO_4)_2$. A small peak around 8980 eV can be found in the ash sample of 10% co-incineration, which also can be found in the characteristic peaks of CuO. However, in the ash of 20% and 50%, its characteristic peak is similar to the ash of sludge and $Cu_3(PO_4)_2$.

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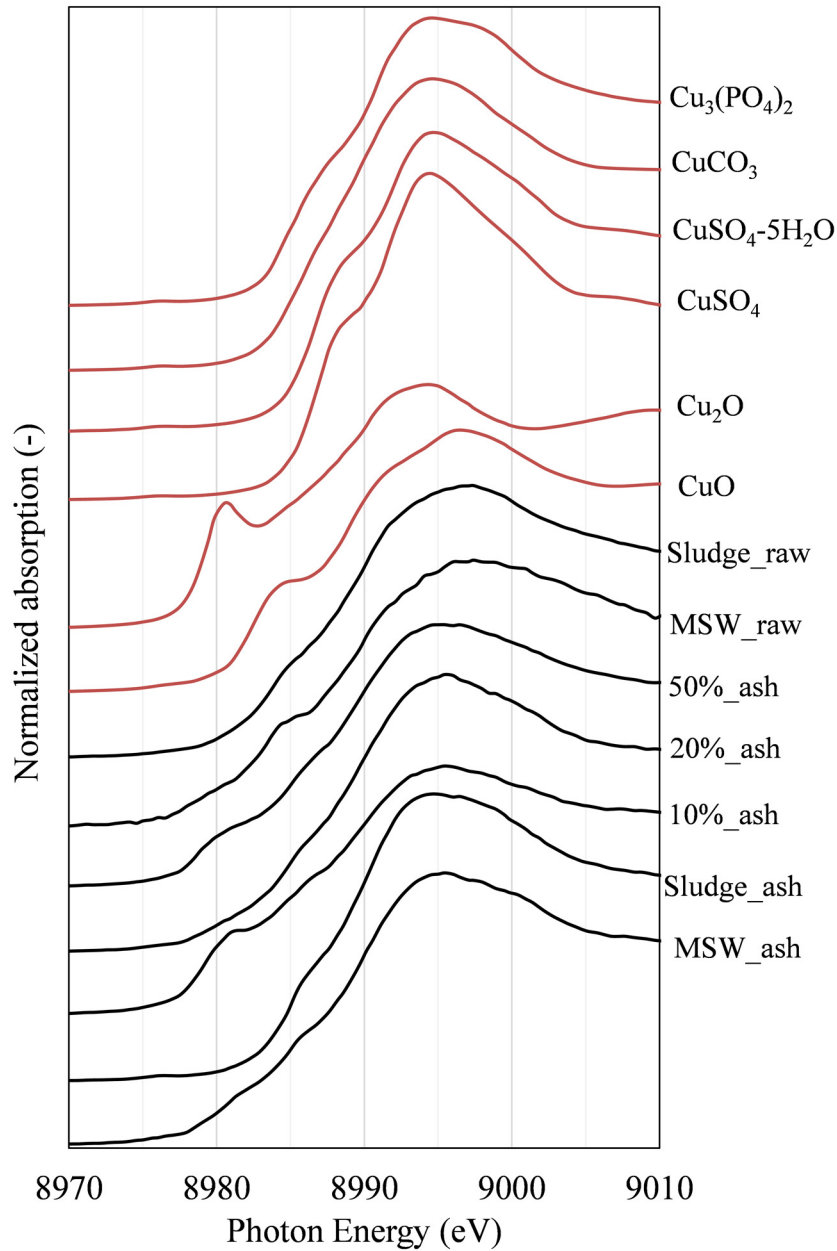
Figure 3.6 XAFS spectra of Cu standard substances and samples.

However, the differences in XAFS are both obvious for Cu, to make a more accurate evaluation, a linear combination fitting (LCF) was performed in the range of 8970 – 9010 eV. In addition, the residual value (R) was calculated using Eq. (3.6) to evaluate the fitting result which is shown in Table 3.3.

$$R = \sum(\text{XAFS}_{\text{sample}} - \text{XAFS}_{\text{standard}})^2 / \sum(\text{XAFS}_{\text{sample}})^2 \qquad (3.6)$$

The lower the R value, the smaller the residual, and the closer the fitting results. The R value of raw MSW and sludge are low for CuO which suggests that the chemical state of Cu in raw MSW and sludge is composed mainly of CuO. After mono-incineration, the Cu state is

supported to be CuO in MSW ash, but which is supported to be $Cu_3(PO_4)_2$ in sludge ash. Phosphate has relatively high stability, it can stabilize heavy metals to form metal phosphates [31][32][33], and CuO(s) is unstable because it may form CuO(g) or CuCl(s) and volatilize with high temperature or the presence of Cl [34].

That may explain that the residual ratio of Cu in the mono-incineration of sludge is higher than that in the MSW (Figure 3.5). Moreover, Cu chemical state in 20% and 50% co-incinerated ash are most likely be $Cu_3(PO_4)_2$, and which in 10% is most like be CuO or Cu_2O. This indicates that there was a higher residual ratio of Cu and P at the 20% and 50% blending ratio (Figure 3.5). This confirms that the co-incineration of MSW and sludge increases the Cu residual ratio due to P stabilized Cu to form copper phosphate.

Table 3.4 R values obtained by LCF for the XAFS spectra of each sample.

Sample	CuO	Cu_2O	$CuSO_4$	$CuSO_4-H_2O$	$CuCO_3$	$Cu_3(PO_4)_2$
MSW_ash	0.0029	0.0303	0.0220	0.0121	0.0095	0.0069
Sludge_ash	0.0127	0.0558	0.0072	0.0029	0.0020	0.0005
10%_ash	0.0079	0.0083	0.0566	0.0398	0.0334	0.0311
20%_ash	0.0042	0.0362	0.0165	0.0080	0.0056	0.0039
50%_ash	0.0033	0.0198	0.0313	0.0192	0.0153	0.0013
MSW_raw	0.0031	0.0320	0.0356	0.0219	0.0193	0.0141
Sludge_raw	0.0037	0.0397	0.0200	0.0102	0.0081	0.0047

3.4 Conclusion

In this study, the influence of P content in sludge on heavy metals of sludge and MSW co-incineration was evaluated. The results of TG-DTA showed that MSW had 3 exothermic peaks, while the sludge had only one peak, which means that the incineration of MSW was caused by organic matter and fixed carbon, while it was mainly caused by volatile matter in the sludge. The weight loss peak at 460 °C during co-incineration is a little shifted to the high temperature with the blending ratio of sludge. This indicated that a interaction effect occurred during co-incineration, which delays the pyrolysis and incineration of polymers and fixed carbon. The residual ratio of heavy metals in sludge mono-incineration was higher than that in MSW mono-incineration. It is inferred that the Cl content of MSW (0.757%) is much higher than that of sludge (0.068%), therefore, heavy metals were affected by Cl to form metal chlorides and volatilize. The interaction effect of co-incineration can be seen in Cu, but not in Cd and Pb. The XAFS analysis proved that Cu in the MSW ash exists in the form of CuO(s), but in the form of $Cu_3(PO_4)_2$ in the sludge ash, and $Cu_3(PO_4)_2$ also found in the co-incineration ash. CuO(s) is relatively unstable and may form CuO(g) or CuCl(s) and volatilize with high temperature and the presence of Cl. Therefore, it is known that P in the sludge has a stable effect on Cu during the co-incineration process.

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Chapter 4 Feasibility study on the co-incineration of sewage sludge and municipal solid waste in Taiwan

4.1 Introduction

As the population and economy continue to grow rapidly, improving the quality of life and health becomes essential; environmental issues are thus increasing more attention. Taiwan's sewage treatment rate has increased from 28.4% in 2015 to 37.9% in 2020 [1]. With the increase in the sewage treatment rate, the amount of sewage sludge also increases. Only a small part (<15%) of sewage sludge is incinerated or recycled for agriculture because these treatments are costly. Therefore, most of the sludge with higher water content is still and directly disposed at the landfill site in Taiwan [2]. However, due to the reduction of the landfill volume, the difficulty of constructing new landfills, the treatment of leachate with high organic load, and the diffusion of generated methane (CH_4) as a greenhouse gas (GHG), the effective treatment of sewage sludge has become a very important issue in Taiwan. In 2019, the amount of incinerated municipal solid waste (MSW) is accounted for 72.5% of the total incineration capacity [3], with 27.5% of the remaining capacity in MSW incinerators (MSWIs).

So far, in Japan, urban metabolic facilities such as MSWIs and wastewater treatment plants (WWTPs) with different functions have been operated separately [4], and there has not been much cooperation involving the exchange of substances and energy. However, most sewage sludge in Taiwan is currently disposed of in landfills, with the remainder being co-incinerated with MSW [5]. Moreover, the lower heating value (LHV) of MSW and dewatered sewage sludge in Japan and Taiwan are similar: 9,360 kJ/kg MSW [6], 1,350 kJ/kg sewage sludge (the result of chapter 2), and 9,600 kJ/kg MSW [3], 1,220 kJ/kg sewage sludge [7], respectively. Therefore, co-incineration could be easily promoted in Taiwan as well as in Japan.

Takaoka et al. [4] calculated the heat and mass balance in a model WWTP and MSWI. They indicated that co-digestion of kitchen waste and sewage sludge, and the co-incineration of kitchen waste and sewage sludge reduced the life cycle cost by 30%, energy consumption by 54%, and GHG emissions by 41%. Matsuo et al. [8] established the database of MSWIs and WWTPs in Japan, analyzed all combinations between MSWI and WWTP, and screen the cooperative combination under some necessary conditions for co-incineration of sewage sludge and MSW. Then, they calculated the potential for co-incineration between WWTPs and MSWIs in Japan and determined that 367 combinations between WWTPs and MSWIs met the necessary conditions for co-incineration. This means that 15.3% of the total sewage sludge generated annually in Japan can be treated.

Hence, considering the characteristics of sewage sludge and the remaining capacity of MSWIs in Taiwan, the co-incineration of sewage sludge and MSW might be one of the development directions of waste management. Whether or not sewage sludge with a high-water content can be co-incinerated with MSW without adding auxiliary fuel seems to depend on the water content of sewage sludge and the mixing ratio between sewage sludge and MSW. In addition,

the remaining capacity of MSWI and the distance between WWTP and MSWI where sewage sludge is transported may also be constraints.

However, the necessary conditions for practical and efficient co-incineration of sewage sludge and MSW, including the distance, the remaining capacity, and the mixing ratio are not clear in Taiwan. In addition, it is necessary to determine the maximum amount of sludge as a potential that can be co-incinerated with MSW for the future waste management plan. Finally, it must be clarified to what extent GHG emissions can be reduced by co-incineration in Taiwan. Therefore, this study aims to elucidate the necessary conditions, potential, and GHG emission of sewage sludge and MSW co-incineration by current operating conditions and statistics of WWTPs and MSWIs in Taiwan. Currently, Taiwan has only one plan to build a new MSWI in near future, so this study only investigated and analyzed the current situation in Taiwan. The flowchart of this research is shown in Figure 4.1. After data collection, the necessary conditions: the remaining capacity (the operating ratio), the mixing ratio, and the distance were set to filter out the efficient combinations of both facilities which were based on the existing literature, statistics, Google Maps API, and the operation data of each facility. Using these necessary conditions, 4 sludge treatment scenarios were analyzed to screen possible combinations of WWTP and MSWI and calculate the potential of co-incineration which is including the number of combinations and treatable sludge amount in all combinations. Finally, GHG emissions of co-incineration were also being evaluated.
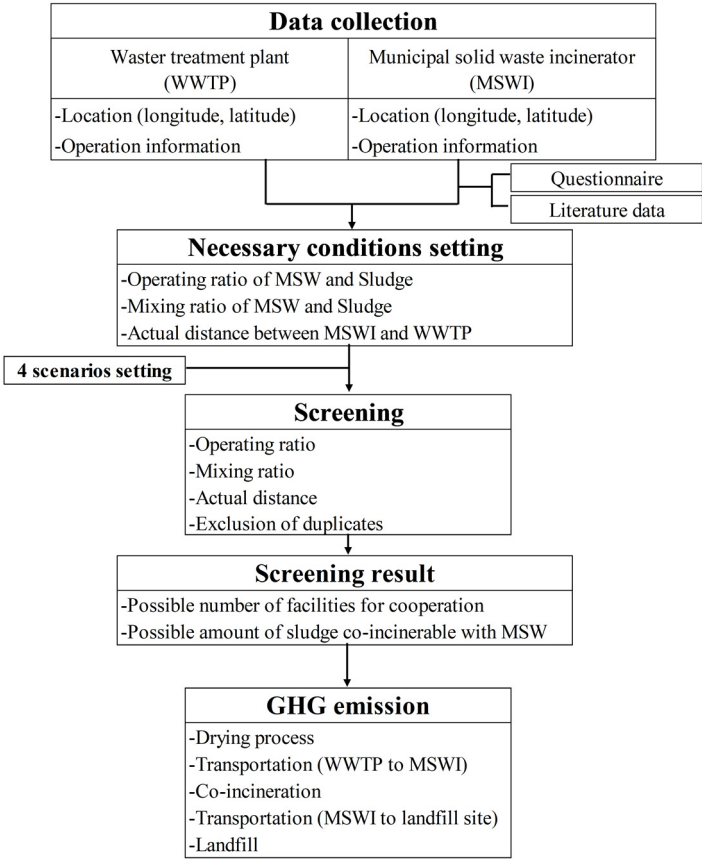
Figure 4.1 Flowchart of this research.

4.2 Survey methods and necessary conditions

4.2.1 Database

The survey database items are listed in Table 4.1. The basic information of WWTPs and MSWIs in 2019 was obtained from the database of the National Statistics of the Environmental Protection Administration in Taiwan [3] and the Sewage System Office, Construction and Planning Agency, Ministry of the Interior in Taiwan [9]. In addition, we referred to the survey result from the Taiwanese Government which had included a sewage sludge reduction plan and WWTP management plan [7].

Table 4.1 Survey database items.

MSWI	WWTP
City	City
Name	Name
Location (longitude, latitude)	Location (longitude, latitude)
Commencement year	Planned treatment (m^3/day)
Design capacity (tons/day)	Actual treatment (tons/year)
Collection source (MSW, industrial waste)	Industrial wastewater (m^3/min)
Incinerated quantity (tons/year)	Electricity used (kWh/year)
Electricity generating quantity (kWh/year)	Dewatered sludge generated (wet-basis tons/year)
Electricity selling quantity (kWh/year)	The moisture of dewatered sludge (%)
Bottom ash (tons/year)	Dried sludge generation (tons/year)
Fly ash (tons/year)	The moisture of dried sludge (%)
Operating ratio (%)	Drying process
	Drying method
	Final sludge treatment

We also conducted a questionnaire survey on the 24 MSWIs to understand the actual co-incineration status. As shown in Figure 4.2, by importing this information into Google My Map, this established database can provide the location, operation ratio, and treat the amount of MSW in MSWI, sewage sludge generation amount in WWTP in visual. Solid squares and solid circles show the locations of MSWIs and WWTPs, respectively. Circled WWTPs will be explained in 4.3.1. Moreover, using this database, the actual distance between MSWI and WWTP can be calculated. Totally 24 MSWIs and 64 WWTPs are operating in Taiwan's mainland. In 2019, the treated amount of MSW is approximately 6.53 million tons in 24 MSWIs. Among the 64 WWTPs, 14 of which do not have sludge information, so we excluded these 14 WWTPs from this study. The other 50 WWTPs generated 53,800 tons of dewatered sludge with an average moisture content of 75% and 3,850 tons of dried sludge with a moisture content of 10~40%.

MSWI	
Name	Jhunan plant
Location	24.6731, 120.83571
Commencement year	2008
Design capacity	500 ton/day
Collection source	MSW 151,000 ton/year
	Industrial waste 13,500 ton/year
Incinerated quantity	164,000 ton/year
Electricity generation	92,800 kWh/year
Electricity sell	76,000 kWh/year
Bottom ash generation	20,600 ton/year
Fly ash generation	7,490 ton/year
Operating ratio	89.90%

WWTP	
Name	Puzih
Location	23.4654,120.2300
Planned treatment	4,200 m^3/day
Actual treatment	1,790 ton/year
Electricity used	243 kWh/year
Dewatered sludge generation	121 ton/year
Dewatered sludge moisture	78.60%
Dried sludge generation	-
Drying process	Constructing

Figure 4.2 Location of 24 MSWIs and 50 WWTPs in Taiwan.

4.2.2 Current co-incineration status of sewage sludge and MSW in Taiwan

The dried sludge from a few WWTPs has been already co-incinerated with MSW in some MSWIs. The actual co-incineration situation obtained from the established database and the results of the questionnaire survey (Figure A4) were used as a reference for determining the necessary conditions. The situation of the co-incineration in Taiwan is shown in Table 4.2 and explained below.

4.2.2.1 Sludge drying and anaerobic digestion

In response to the problem of sludge treatment, the Taiwanese government proposed a sludge reduction plan to subsidize the installation of sludge drying equipment in WWTPs with stable sludge production. The goal is to reduce the moisture content of sludge from 80% to 30% and promote the dry sludge to be sent to the MSWIs for co-incineration. Among the 50 WWTPs, 11 WWTPs (drying process operating in Table 4.2) had implemented the drying process, the dried sludge of 2 WWTPs is transported to 4 MSWIs for co-incineration. The dried sludge of 9 WWTPs from 11 WWTPs was not co-incinerated but treated in some private companies. These 11 WWTPs produced both dewatered sludge and dried sludge as the final product. In addition, other 8 WWTPs (drying process constructing in Table 4.2) had applied for government subsidies to implement the drying process, although they have not yet been activated. Anaerobic digestion has been installed to only 10 WWTPs of 50 WWTPs. However, the generation amount of biogas and their utilization in these 10 WWTPs were unknown. Therefore, anaerobic digestion was not covered in this research.

Table 4.2 Current co-incineration status of sewage sludge and MSW in Taiwan.

Status	No	Name	Dewatered sludge Generation (ton/year)	Moisture (%)	Dried sludge Generation (ton/year)	Moisture (%)	No	Name	Planning treatment (ton/day)	Actual treatment (ton/year)	Operating ratio (%)	Distance (km)	Mixing ratio (%)	Operating ratio (%)
										WWTP		MSWI	Calculated value for necessary conditions	
Drying process operating	1	Dihua	-	79.4	1930	10	1	Beitou	1,800	473,000	72.0	5.61	unknown	unknown
							2	Neihu	900	152,000	46.3	12.3	unknown	unknown
							3	Muzha	1,500	182,000	33.2	15.5	unknown	unknown
	2	Danshui	2,200	77.5	494	15	4	Bali	1,350	419,000	85.0	25.6	0.52	85.5
	3	Yilan	1,150	79.3	84	30	-	-	-	-	-	-	-	-
	4	Luodong	41	78.3	486	30	-	-	-	-	-	-	-	-
	5	Zhudong	193	77.3	203	10	-	-	-	-	-	-	-	-
	6	Hualien	-	66.8	442	30	-	-	-	-	-	-	-	-
	7	Toufen	325	78.2	1	30	-	-	-	-	-	-	-	-
	8	Chiayi-large	109	77.9	11	30	-	-	-	-	-	-	-	-
	9	Anping	2,050	78.5	40	40	-	-	-	-	-	-	-	-
	10	Nanzi	2,830	78.4	161	20	-	-	-	-	-	-	-	-
	11	Fengshanxi	4,980	57.8	-	30	-	-	-	-	-	-	-	-
Drying process constructing	12	Bali	9,430	71.8	3800*	30	4	Bali	1,350	419,000	85.0	8.67	0.92	85.8
	13	Linkou	2,100	78.4	649*	30	4	Bali	1,350	419,000	85.0	12.8	0.17	85.2
	14	Guishan	1,320	79.6	385*	30	5	Taoyuan	1,350	427,000	86.7	21.0	0.11	86.8
	15	Keya	2,700	79.7	782*	30	6	Hsinchu	900	237,000	72.1	6.78	0.36	72.4
	16	Futian	2,680	75.1	953*	30	7	Wuri	900	293,000	89.2	7.64	0.35	89.5
	17	Douliu	715	79.7	207*	30	-	-	-	-	-	-	-	-
	18	Puzih	121	78.6	37*	30	-	-	-	-	-	-	-	-
	19	Liukuaicuo	2,230	79.5	655*	30	8	Kanding	900	251,000	76.4	24.0	0.29	76.6

*the amount was calculated as the moisture of 30%

4.2.2.2 Co-incineration of sewage sludge and MSW

Currently, although 2 WWTPs and 4 MSWIs engage in co-incineration in Table 4.2. Part of the 1,930 tons/year of sludge produced in Dihua WWTP (No.1) was sent to 3 MSWI for co-incineration, but the amount and ratio of sludge are unknown. All dewatered sludge: 2,200 tons/year and dried sludge:494 tons/year produced in Danshui WWTP (No.2) was sent to Bali plant MSWI (No.4). Table 4.2 also shows the data of 8 WWTPs under construction of the drying process, 6 WWTPs of them have already planned the co-incineration of sludge, and the cooperative MSWI also has been confirmed. The other 2 WWTPs currently do not have plans for the co-incineration of sludge. Since the drying process has not operated yet, the amount of dried sludge (* marked) was calculated as moisture content of 30% from the data of dewatered moisture content and production.

Based on this information, we calculated the necessary conditions of the actual distance between WWTP and MSWI, mixing ratio, and operating ratio. Detailed calculating equations will be further explained in 4.2.3.

4.2.3 Necessary conditions for co-incineration

Not all WWTPs and MSWIs are capable of co-incineration unconditionally. The distance between WWTP and MSWI needs to be close, the MSWI must have the capacity to accept sludge, and the co-incineration of sludge does not worsen the combustion conditions of the MSWI.

We assumed that sludge is transported directly from WWTPs to MSWIs by vehicles and suppose that all sludge in one WWTP is transported to MSWI, and to co-incinerate, the actual distance between WWTP and MSWI must be considered. For the stable operation of MSWI, operating and mixing ratios of the co-incineration should be considered. Therefore, necessary conditions should be established to ensure that the combinations are efficient and feasible.

4.2.3.1 Operating ratio of sewage sludge and MSW

If a MSWI is to receive sewage sludge, the total amounts of MSW and additional sewage sludge incinerated must be less than the incineration capacity of MSWI. The operating ratio is defined by Eq. (4.1).

$$R_{op} = \frac{M_S + M_{wa}}{C_{wa}} \times 100\% \qquad (4.1)$$

where R_{op} represents the operating ratio (%), C_{wa} is incineration capacity (ton/year) of MSWI, M_s is the sludge mass (ton/year), and M_{wa} is the waste mass (ton/year). From Table 4.2, the calculated operating ratios were ranged from 72.4% to 89.5%. Therefore, we set below 90 % as an operating ratio of the necessary condition.

4.2.3.2 Mixing ratio of sewage sludge and MSW

The moisture content of dewatered sludge is generally higher than that of MSW, when the sludge is added, the moisture content will increase, and the internal temperature of the incinerator will fall. To ensure a stable co-incineration, the sludge input must be controlled. The mixing ratio is given by Eq. (4.2).

$$R_m = \frac{M_S}{M_S + M_{wa}} \times 100\% \qquad (4.2)$$

where R_m represents the mixing ratio (%) of sludge and MSW.

From Table 4.2, the calculated mixing ratios in cases of dried sludge were ranged from 0.11% to 0.92%. Therefore, we set below 1.0 % as a mixing ratio of the necessary condition. The dried sludge particle size is about 1~5 mm in Taiwan. Most MSWIs are equipped with the mechanic-grate (Stoker type) in Taiwan. The gap width of the grate is only about 1~3 mm. If the mixing ratio is too high, the dried sludge may drop into the gap of the furnace, blocking the ash collection system, the mixing ratio should be limited to 1.5% [10]. This literature value is consistent with the 1.0 % set from the actual and expected values in Table 4.2, and this 1.0% is the value set on the safe side.

In the case of dewatered sludge, according to the results of the questionnaire survey, Wuri MSWI accepted industrial sludge with 60~80% of water content and performed co-incineration with the mixing ratio is 2~3%. Thus, we set up the mixing ratio of the necessary condition in the case of dewatered sludge to 3.0% based on practical experience and facility data of the questionnaire survey. The median of mixing ratio in Japan according to a Japanese questionnaire survey was approximately 6.0% [6]. This value is comparatively higher than that in Taiwan. Two possible reasons were speculated, first, for Taiwan itself, since Taiwan's sludge production is relatively small compared with the amount of MSW cause a low mixing ratio. Second, in terms of public sewage rate, compared with Japan's 80% [11], Taiwan only has 38%, so this may also be the reason why Japan's sludge production and mixing ratio are higher than that in Taiwan.

4.2.3.3 Actual distance between WWTPs and MSWIs

Since transportation is carried by vehicles, the actual distance is considered to be an important factor. Google Maps API (http://code.google.com/apis/maps/) was used to calculate the actual distance between WWTPs and MSWIs. The actual distance refers to the distance traveled by the vehicle during transportation along the road. The actual traveling distance between facilities with experience versus facilities under the planning of co-incineration ranged from 5.6 km to 25.6 km from Table 4.2, respectively. Therefore, we set up the distance criterion as 30 km.

4.2.3.4 Exclusion of duplicates

One MSWI may be in cooperation with more than one WWTP, however, if one WWTP simultaneously sends sludge to the multiple MSWIs, the amount of sludge sent to each MSWI may not be grasped, resulting in an unclear mixing ratio and operating ratio. To avoid such a complicated situation, we assumed that WWTPs would only cooperate with one MSWI under the condition of the minimum actual distance, the possible combinations were WWTP: MSWI at a 1:1 ratio, or several WWTPs to 1 MSWI.

4.2.4 Scenarios setting for co-incineration

According to the government policy, the sludge discharged from the WWTP includes dewatered and dried sludge, and it is estimated that the amount of dried sludge will increase in the future. We have established four scenarios to calculate the co-incineration potential of dewatered and dried sludge, which is shown in Table 4.3.

Table 4.3 Scenario simulations by different sludge status and moisture content.

	Simulation	Number of WWTPs	Sludge status	Moisture
Scenario 1	All WWTPs without drying equipment	50	Dewatered sludge	36.7-82.1%
Scenario 2	Current situation	11	Dried sludge -current	10-40%
		39 (49)	Dewatered sludge	36.7-82.1%
Scenario 3	Additional drying equipment planned	11	Dried sludge -current	10-40%
		8	Dried sludge -planned	30%
		31	Dewatered sludge	36.7-82.1%
Scenario 4	All WWTPs with drying equipment	11	Dried sludge -current	10-40%
		8	Dried sludge -planned	30%
		31	Dried sludge -future	30%

Scenario 1 (S1) assumed that all 50 WWTPs have no drying process in place; all sludge emitted as dewatered sludge with its original moisture level. Scenario 2 (S2) simulated the current situation in Taiwan: 11 WWTPs produced dried sludge with moisture content 10~40%, and 39 WWTPs produced dewatered sludge with moisture content 36.7~82.1%; although 11 WWTPs were equipped with drying equipment, but dewatered sludge was still being produced in 10 WWTPs, the dewatered sludge and dried sludge produced by the same WWTP were then calculated separately based on their respective amount and moisture content. Scenario 3 (S3) assumed that additional 8 WWTPs were planning to implement the drying process with the projected moisture content of 30%, and 31 WWTPs produced dewatered sludge with moisture content 36.7~82.1%. Scenario 4 (S4) simulated that all 50 WWTPs engaged in the drying process. In all scenarios except for S2, the volume reduction of dewatered sludge by the drying process and vice versa was expressed as an Eq. (4.3).

$$\frac{V_1}{V_2} = \frac{(100-p_2)}{(100-p_1)} \qquad (4.3)$$

V_1 represents the sludge volume when the sludge moisture content is p_1, and V_2 is the sludge volume when the moisture content is p_2.

4.3 Results and discussion

4.3.1 Screening results

Initially, there were a total of 1,200 co-incineration combinations of 24 MSWIs and 50 WWTPs. Then calculated the necessary conditions (operating ratio, mixing ratio, and distance) for each of 1,200 combinations, and screen by the upper limit set and exclusion of duplicates in 4.2.3.1 to 4.2.3.4).

After the screening of (1) the operating ratio below 90%, (2) the mixing ratio below 3% for dewatered sludge and 1% for dried sludge, (3) the actual distance within 30km, and (4) exclusion of duplicates in sequence, the screening results of cooperative combination number for each scenario are shown Table 4.4. There are 39 combinations including 39 WWTPs and 18 or 19 MSWIs of screening results for all scenarios, which means the drying process does not affect the combinations that can be co-incineration. The 39 WWTPs in all scenarios are the same WWTP, but there is one more MSWI in S2~S4 than in S1. The reason is that, in the case of S1, if the dewatered sludge produced by one of the WWTP is sent to neighboring MSWI itself has a higher operating ratio (89.9%). To avoid the operating ratio of co-incineration exceeding 90%, it needs to be sent to another MSWI. However, in the case of S2–S4, the amount of dried sludge is only 1 ton, which is an extremely small amount for the MSWI, and the co-incineration will not affect the operating ratio.

Table 4.4 Combination number of screening results.

Screening result	Scenario 1		Scenario 2		Scenario 3		Scenario 4	
Initial combination number	1200		1200		1200		1200	
Final combination number	39		39		39		39	
Facility numbers	WWTP	MSWI	WWTP	MSWI	WWTP	MSWI	WWTP	MSWI
	39	18	39	19	39	19	39	19

Figure 4.3 shows the change in the amount of co-incinerable sludge during the screening process. The screening of operating and mixing ratio did not change the amount of co-incinerable sludge, only distance affected it. The same trend can be seen in every scenario. Therefore, distance is an important factor affecting the feasibility of co-incineration. Although the amount of co-incinerable sludge for S1, S2, S3, and S4 is 92%, 92%, 83%, and 89% of the national sludge generation, respectively, but its co-incinerable amount is the same at 89.7% on a dry basis. The survey in Japan [8] shows that when operating ratio < 83%, mixing ratio < 18%, and the distance between WWTP and MSWI < 18km, 15.3% of the total sewage sludge generation can be co-incineration in Japan. Compared with Japan, sludge co-incineration potential is much greater. In addition to the different necessary conditions setting, the main reason is considered to be that the sludge amount is less, so MSWI is capable of receiving most

of the sludge in Taiwan. The 11 WWTPs that cannot be co-operated are green marked in Figure 4.2. Among them, 7 WWTPs do not have MSWI within 30km, and 4 WWTPs that neighboring MSWI itself has an operating ratio higher than 90%.
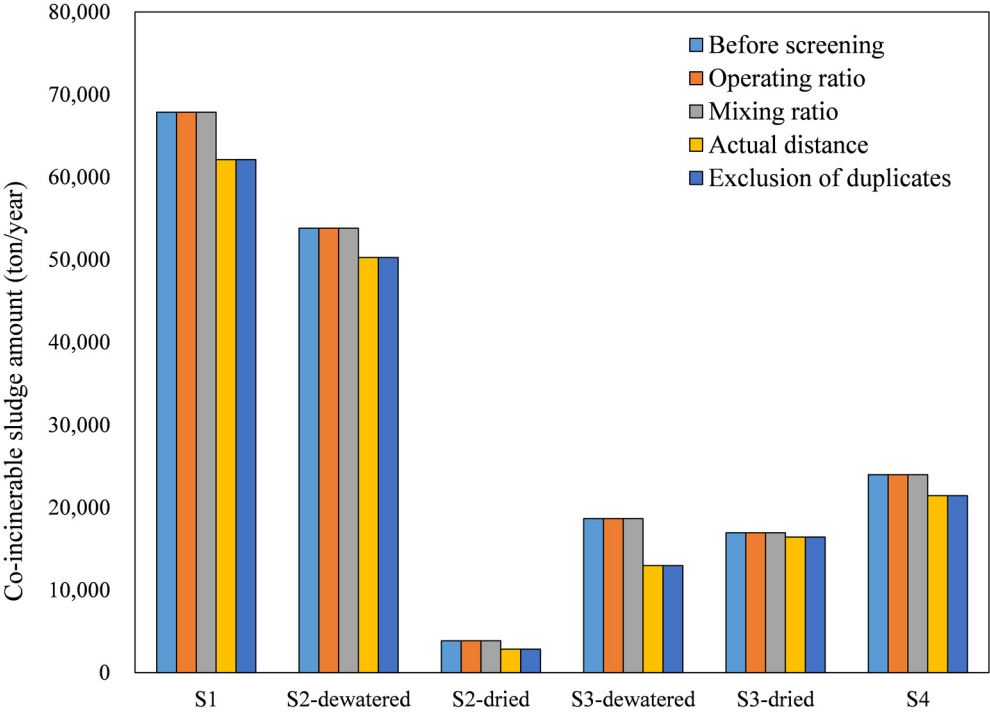
Figure 4.3 The screening result of sludge co-incinerable amount.

4.3.2 GHG emission

As shown above, whether the sludge is dried or dewatered does not affect the potential for co-incineration. However, The moisture content and amount of sludge will influent the transportation from WWTP to MSWI and GHG emissions. In addition, scenario 0 (S0) was added to compare GHG emissions when dewatered sludge was disposed to landfill directly without other treatment.

The boundaries of the GHG emissions are shown in Figure 4.4. The calculation of GHG emissions includes the process of drying, transportation, co-incineration, and landfill. All WWTPs engage in the dewatering process, so we did not include it in the calculation of GHG emissions boundaries. Each GHG emission factor was converted to the CO_2-equivalent (CO_2-eq) based on its global warming potential (GWP). The GWPs for CH_4 and N_2O are equivalent to 25 and 298 tons of CO_2 (ton CO_2-eq), respectively [12]. The coefficients required for GHG calculations are listed in Table 4.5. The detailed description is as follows.
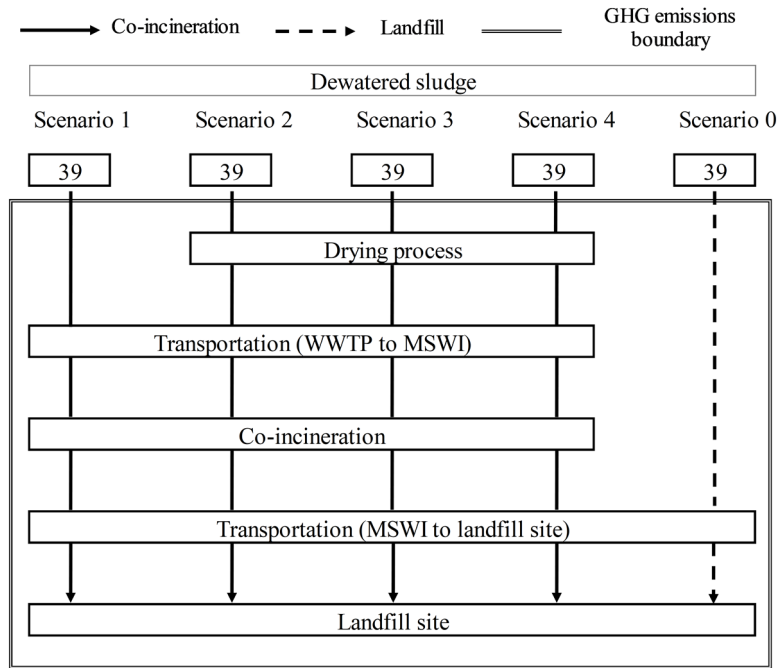
Figure 4.4 Boundaries of GHG emissions.

Table 4.5 GHG emission factors of the various processes and associated energy consumption.

Energy	Fuel	Electricity	CO_2 (ton CO_2/ton)	CH_4 (ton CH_4/ton)	N_2O (ton N_2O/ton)	GHG emissions	Unit	Note
Fuel			1.879	0.000033	0.000003	1.88	kg CO_2-eq/m^3	Indirectly emission
Electricity						0.509	kg CO_2-eq/kWh	Indirectly emission
Treatment								
Drying								
Indirect heating	0.178	0.0136				0.342	ton CO_2-eq/ton	Remove 1 ton water
Freeze drying	0	0.361				0.184	ton CO_2-eq/ton	Remove 1 ton water
Transportation								
Trucking						0.00024	ton CO_2-eq/ton-km	Capacity: 4.5 tons
Co-incineration								
Sludge				0.0000015	0.000263	0.0784	ton CO_2-eq/ton	30% moisture
Landfill								
Operation of landfill						0.0379	ton CO_2-eq/ton	Indirectly emission
Sludge				0.133		3.325	ton CO_2-eq/ton	
Incinerated ash				0		0	ton CO_2-eq/ton	

4.3.2.1 GHG emissions during the drying process

In Taiwan, there are two types of dryers currently used in WWTPs: indirect heating and freeze-drying. The calculations of the drying process are based on the data of the indirect heating dryer in WWTP No.1 [13] and the freeze-drying dryer in WWTP No.6 [14] (Table 4.2). The indirect heating dryer of WWTP No.1 uses electricity, biogas, and natural gas as energy sources. To remove 1 kg of water from sludge by indirect heating dryer requires 0.178 m^3 of natural gas and 0.0136 kWh of electricity. For the freeze-drying dryer, the total power consumption divided by the total amount of water removed shows that the electricity required to remove 1 kg of water from sludge is 0.361 kWh. The GHG emission coefficient of electricity was 0.509 kg CO_2-eq/kWh, and that of natural gas was 1.879 kg CO_2/m^3, 0.000033 kg CH_4/m^3, and 0.000003 kg N_2O/m^3 in 2019 in Taiwan [15]. Thus, the GHG emission amount associated with the removal of 1 ton of water in sludge via indirect heating dryer was calculated as 0.342 ton CO_2-eq, and that of the freeze-drying dryer was calculated as 0.184 ton CO_2-eq. We first calculated the amounts of water removed from sludge by drying in S2, S3, and S4 to the set moisture content (30%) and multiplied these by the relevant GHG coefficients to obtain the total GHG emissions.

4.3.2.2 GHG emissions during transportation

The vehicle for transportation assumed in this study is a truck for business with a capacity of 4.5 tons. According to the carbon footprint emission factors of the Environmental Protection Agency of Taiwan [16], the GHG emission of the truck for business is 0.00024 ton CO_2-eq/ton-km (Carrying 1 ton and running 1 km emits 0.00024 ton of CO_2, and this value includes CO_2 emission from fuel consumption). Transportation emissions (transport the sludge from WWTP to MSWIs or the ash from MSWI to a landfill site) were calculated for each scenario using Eq. (4.4).

$$GHG_{transportation} = \left(\frac{M}{4.5}\right) \times \left(0.00024 \times D \times (8.8 + 4.3)\right) \quad (4.4)$$

where $GHG_{transportation}$ is the GHG emission (ton CO_2-eq/year) during transportation, M is the mass (ton/year) of sludge or ash, and D is the distance (km) between the WWTP and MSWI or MSWI and landfill site. The information on the landfill site referred to government statistics [17]. The number of trips/year is calculated by dividing the total sludge amount by 4.5 tons (the amount transported/trip) multiply by loads (inbound: 8.8 tons, outbound: 4.3 tons [empty truck]) and the distance between the facilities, and then multiplying by the GHG emissions coefficient of 0.00024 ton CO_2-eq/ton-km.

4.3.2.3 GHG emissions from co-incineration

GHG emissions from incineration of MSW only were not included in the calculation from S1 to S4, assuming that they did not depend on co-incineration with sludge. Namely, the GHG emissions at MSWI were limited to sludge combustion in the calculation. The GHG emissions of sludge are based on the "National Greenhouse Gas Inventory Report of JAPAN 2021" [18]. As sludge is biomass, the CO_2 emitted on incineration is not included in the calculation of GHG emissions; the GHG emission coefficients of sludge are 0.0000015 ton CH_4/ton and 0.000263 ton N_2O/ton from sludge mono-incineration. Therefore, the emission coefficient for sludge incineration was calculated as 0.0784 ton CO_2-eq/ton. In addition, the heat generated during incineration can be converted into electricity, which can offset GHG emissions. We first calculated the energy conversion efficiency of each MSWI based on the amount of MSW incinerated (M_{MSW}, ton/year), electricity generation (Q_{EG}, kWh/year), and the LHV of MSW, as shown in Eq. (4.5).

$$ECE = \frac{Q_{EG} \times 3600}{LHV \times M_{MSW} \times 1000} \qquad (4.5)$$

where ECE is the energy conversion efficiency (%) of MSWI. The 3,600 kJ is the energy of 1 kW of electricity, and the average LHV of MSW is 9,600 kJ/kg [3] in Taiwan.

The electricity generated by sludge can be calculated from the MSWI conversion efficiency. The heating value of sludge varies by moisture content; the average higher heating value (HHV) in Taiwan is approximately 14,000 kJ/kg including anaerobically digested sludge, and the average hydrogen (H) content is about 5%TS [8]. Therefore, the LHV of sludge on a dry basis (LHV_d) can be calculated by Eq. (4.6), and the LHVw (LHV of sludge with wet basis) is given by Eq. (4.7).

$$LHV_d = HHV - 4.186 \times 600 \times 9 \times H \qquad (4.6)$$

$$LHV_w = \frac{100-W}{100} \times LHV_d - 4.186 \times 600 \times \left(\frac{W}{100}\right) \qquad (4.7)$$

where H is the hydrogen concentration (% dry base), and W is the moisture content (%) of dewatered sludge.

The GHG emissions on co-incineration are calculated by Eqs. (4.8)~(4.10).

$$GHG_{CO} = GHG_{In-sludge} + GHG_{EC} - GHG_{EG} \qquad (4.8)$$

$$GHG_{In-sludge} = M_s \times 0.0784 \qquad (4.9)$$

$$GHG_{EC\,(EG)} = Q_{EC\,(EG)} \times 0.509 \qquad (4.10)$$

where GHG_{CO} is the GHG emissions of co-incineration, $GHG_{In\text{-}sludge}$ is the GHG emissions of sludge incineration, GHG_{EC} is the GHG emissions of electricity consumption, GHG_{EG} is the offset GHG emissions of electricity generation, and Q_{EC} is the quantity of electricity consumption (kWh/year).

4.3.2.4 GHG emissions from landfill

GHG emissions of the landfill are divided into indirect emissions caused by landfill operations and direct emissions reflecting landfill waste decomposition. The indirect emission is 0.0379 ton CO_2-eq/ton [19], and the direct emission from landfilled sludge is 0.133 ton CH_4/ton [20] (anaerobic landfill), which is converted into a GHG emission factor of 3.325 ton CO_2-eq/ton. The final product of S1~S4 is ash, which has no decomposable components, so it only causes indirect emissions without direct GHG emissions during landfilling. The average ash content of sludge is about 43% (dry basis) in Taiwan [7], and the landfill ash level after incinerating is calculated based on the mass of the sludge which was converted to a dry basis.

4.3.2.5 Comparison of the total emissions of the five scenarios

The total GHG emissions (as CO_2 equivalent) of five scenarios are shown in Figure 4.5. GHG emissions of S0 are about 5~8 times to other scenarios which involve direct landfilling of dewatered sludge, and most GHG emissions are caused by methane during the landfill process. The GHG emissions from the drying process increase with the increase of WWTP with drying equipment. The drying of sludge can reduce the GHG emission of transportation and incineration, and the generated electricity can also offset the GHG emission. However, the GHG emission of the drying process is still greater than the indirect reduction in other processes derived from the drying process.

After all, the greatest impact on GHG emission is the energy consumption required for sludge drying, especially water evaporation. At present, sludge is dried in WWTP and therefore requires a lot of energy. Therefore, dewatered sludge (S1) is the best scenario for co-incineration with MSW from the point of view of GHG emissions. Although biogas from the anaerobic digestion process could be a promising heat source for sludge drying in WWTP, the anaerobic digestion process would decrease the HHV of sludge as an energy source in MSWI. However, if the sludge drying process is installed in MSWI and could use the incineration waste heat for sludge drying, the GHG emissions generated by the drying process can be greatly reduced and the power generation may be increased. In this case, it must be transported as dewatered sludge, but GHG emissions from transportation are small from Figure 4.5, and it is considered that there is no significant effect on the whole.
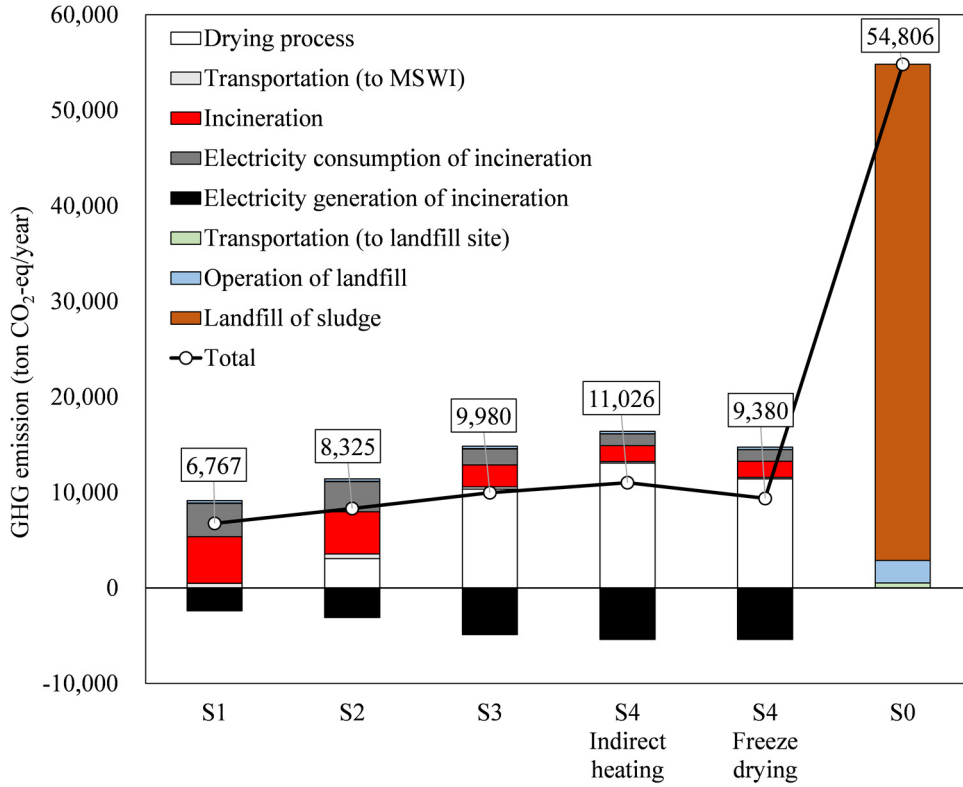
Figure 4.5 GHG emissions of the five scenarios.

4.3.2.6 Calculation of GHG emissions of drying equipment construction

The GHG emissions calculated above are for the steady state. To evaluate the impact of drying equipment installation on subsequent emissions, we calculated GHG emissions during installation using a simplified IO method [21], which calculates GHG emissions by construction costs. The construction costs are divided into civil construction costs ($Cost_{civil}$) and mechanical and electrical costs ($Cost_{me}$) in Japanese yen. First, the Guideline of Sewage Sludge Energy Conversion Technical [22] was used to calculate the costs of civil construction, machinery, and electricity, based on the performance of drying equipment which is employed, which shown in Eq. (4.11)~(4.12).

$$Cost_{civil} = 64.741x^{0.391} \qquad (4.11)$$
$$Cost_{me} = 228.55x^{0.4974} \qquad (4.12)$$

Where x represents the sludge drying capacity of equipment (ton-wet/day).

After obtaining the cost, which was converted into GHG emissions by the emission factor ($I-A$)$^{-1}$ types based on Embodied Energy and Emission Intensity Data (3EID) for Japan using input-output tables (IOTs) [23][24]. This yielded the GHG emissions based on construction costs shown in Table 4.6. The GHG emissions during drying equipment installation, and over the subsequent years of operation are shown in Figure 4.6.

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Table 4.6 Emission factor $(I-A)^{-1}$ values associated with civil construction, and machinery and electricity operations [23][24].

GHG emission factor $(I-A)^{-1}$	ton CO_2eq/million Japanese yen
Civil construction	3.793
Machinery and electricity	3.054

In the constructing year (operation year of 0), S0 and S1 did not have the construction of drying equipment, and its GHG emissions are 0; S2 includes 7 sets of drying equipment, with GHG emissions of 6,767 ton CO_2-eq; S3 includes 16 sets with emissions of 38,141 ton CO_2-eq/year; S4 indirect heating and S4 freeze-drying both include 35 sets with emissions of 55,277 ton CO_2-eq/year. The GHG emissions associated with construction have a large impact (S4 > S3 > S2 > S1 = S0). Overall, from the long-term perspective, direct co-incineration of dewatered sludge has the least emissions.
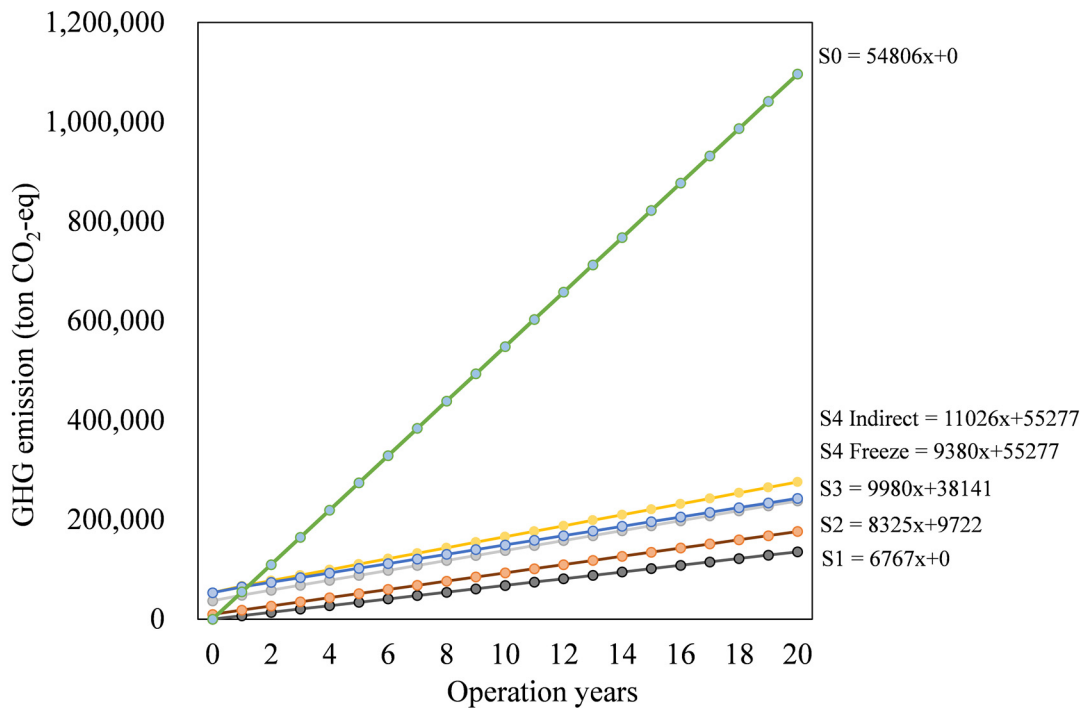

Figure 4.6 GHG emissions during drying equipment installation, and over the subsequent years of operation.

4.4 Conclusion

This study established a database of WWTPs and MSWIs in Taiwan and set the necessary conditions and 4 scenarios to evaluate the number of WWTPs and MSWIs that can be cooperated, the amount of sludge that can be co-incinerated. Finally, GHG emissions were calculated in 5 scenarios. The results obtained were as follows.

(1) A database of WWTPs and MSWIs in Taiwan was established from various statistical data and references. The location information and various facility information can be confirmed, and the actual distance between facilities can also be calculated.

(2) Based on the government's policy direction, information in the database, and questionnaire survey, we calculated and set the Taiwanese necessary conditions of co-incineration: operating ratio below 90%, mixing ratio below 3% for dewatered sludge, and 1% for dried sludge, actual distance between WWTP and MSWI within 30 km.

(3) 4 scenarios were set to evaluate the feasibility of dewatered and dried sludge on the co-incineration. The results of screening from 1,200 combinations between WWTP and MSWI show that all scenarios have 39 combinations extracted.

(4) Regardless of dewatered or dried sludge, the co-incinerable amount is the same on a dry basis, which accounts for 89.7% of national sludge generation. Since the amount of sludge is currently less compared to MSW, the operating ratio and mixing ratio hardly affect the feasibility of co-incineration. Only the distance between WWTP and MSWI as a key factor affects it.

(5) GHG emissions of S0 (direct landfill of dewatered sludge) are about 5~8 times to other 4 scenarios. Although dried sludge can reduce GHG emissions from transportation and incineration, the generated electricity can also offset GHG emissions. However, the GHG emissions of dried sludge are more than dewatered sludge due to the drying process requires more energy. From the perspective of GHG emissions, dewatered sludge (S1) is the best scenario for co-incineration with MSW.

(6) Given the GHG emissions at on initiation of drying equipment construction, from a long-term perspective, the direct co-incineration of dewatered sludge (S1) has the smallest GHG emissions.

(7) We provided the information on the combination of each scenario (including the name of facilities, actual distance, mixing ratio, and operating ratio of WWTP/MSWI); the landfill site closest to the MSWI for final disposal of ash are also listed in Table A2.1-A2.5; hoping this information can provide a reference for the co-incineration of sewage sludge and MSW in Taiwan.

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Chapter 5 Conclusion and future perspectives

5.1 Conclusion

Co-incineration of sewage sludge and municipal solid waste (MSW) is an efficient and safer treatment, and the waste heat of incineration can be converted into electricity to bring benefits. The composition of MSW is vary with urban development, environment, living habits, management policies, etc., and the composition of sewage sludge is affected by geographic conditions, sewage collection method, treatment processes, and chemical additions, and so on. The variability of sewage sludge and MSW is very large. Chorine (Cl), sulfur (S), phosphorus (P) are considered to affect the migration of heavy metals during incineration. Cl is quite abundant in MSW, while S and P are contained in sewage sludge. Therefore, the impact caused by co-incineration needs to be taken seriously.

In this study, the sewage sludge composition of Japan was investigated, the characteristics were clarified through various statistical analyses, and the influencing factors of sewage sludge composition were proposed to provide the prediction. From Chapter 2, it was found that sewage sludge has a higher heating value and can meet the requirements of BSF-15 (biomass solid fuel, a heating value higher than 15 MJ/kg). Therefore, sewage sludge has considerable advantages as a biofuel. However, heavy metals are the factor of sewage sludge recycling that needs to be carefully evaluated. The addition of P during the incineration process may form high-stability heavy metal phosphates, which can reduce the volatilization of heavy metals. Therefore, this study explored the effect of co-incineration of sewage sludge with high P content and MSW on heavy metals. It was found that the Cl in the MSW increases the volatilization of heavy metals, but the P in the sewage sludge stabilized the copper (Cu), and this effect also be achieved during the co-incineration. Co-incineration is a favorable treatment method of solid waste for countries with a small land area. Taiwan is currently in the stage of trial and research on the co-incineration of sewage sludge and MSW. This study also conducted an evaluation of co-incineration feasibility in Taiwan, which was about 89.7% of national sewage sludge could be co-incineration and the GHG emissions of dewatered sludge co-incineration were lower than that of dried sludge. The main conclusions from each chapter are presented below.

Chapter 2 investigated the composition of 119 sewage sludge from 32 wastewater treatment plants across Japan, although the composition of sewage sludge has a large deviation, the element content of sewage sludge in the United States, Sweden, Switzerland, and Japan have the same tendency. Through Hayashi's quantification method I, it was clarified that sewage collection system, digestion process, the coagulant used for sludge dewatering had significant effects on the composition of the element. Especially the digestion process had a major influence on the organic matter. Seasonal effects and the dewatering device had little influence on elemental concentrations. In the Van Krevelen diagram, by comparing H/C and O/C, it was found that the composition of the sewage sludge was similar to RDF. The content of heavy

metals in the combined system was higher than that in the separated system and was greater in digested sludge than in undigested sludge. The Cu content of sewage sludge in Japan is relatively high, exceeding the limit set by fertilizer. However, the heating value of most sludge is higher than 15 MJ/kg which belongs to BSF-15 in Japan, therefore, the sewage sludge in Japan has great potential as an alternative fuel.

Chapter 3 evaluated the influence of P content in sewage sludge on heavy metals of sewage sludge and MSW co-incineration. The TG-DTA results show that the incineration of MSW was mainly caused by organic matter and fixed carbon, but mainly by the volatile matter in the sewage sludge. During the co-incineration, the peak weight loss at 460°C slightly shifts to high temperature with the different blending ratios of sludge. This indicates that the co-incineration process delays the pyrolysis and incineration of the polymer and fixed carbon. The residual ratio of heavy metals in mono-incineration of sewage sludge was higher than that in MSW. It is inferred that the Cl content in MSW (0.757%) is much higher than that in sewage sludge (0.068%), so heavy metals are formed metal chloride by Cl and volatilized. The interaction effect of co-incineration could be seen in Cu, but not in the lead (Pb) and cadmium (Cd). XAFS analysis proves that Cu in the ash content of MSW exists in the form of $CuO(s)$, but it exists in the form of $Cu_3(PO_4)_2$ in the ash content of the sewage sludge, and $Cu_3(PO_4)_2$ was also found in co-incineration ash. $CuO(s)$ is relatively unstable and may form $CuO(g)$ or $CuCl(s)$ and volatilize under high temperature and Cl presence. Therefore, it can be seen that P in the sewage sludge has a stable effect on Cu in the process of co-incineration.

Chapter 4 evaluated the feasibility of co-incineration of sewage sludge and municipal solid waste in Taiwan. A database of wastewater treatment plants (WWTP) and municipal solid waste incinerators (MSWI) in Taiwan was established. Necessary conditions for co-incineration which including operating ratio < 90%, mixing ratio < 3% for dewatered sludge and 1% for dried sludge, actual distance between WWTP and MSWI within 30km were set according to questionnaires, literature data, and government policy reports of co-incineration. 4 scenarios were set according to the drying equipment in the WWTP and screened by necessary conditions in sequence. 39 feasible combinations had been screened out, and which accounts for 89.7% of national sludge generation could be co-incineration. Since the sludge amount is very small compared to MSW, the operating ratio and mixing ratio hardly affect the co-incineration potential. Only the distance between WWTP and MSWI is a key factor. The direct landfill of dewatered sludge generated a large amount of methane due to the decomposition of organic matter, which caused considerable GHG emissions. The co-incineration of dewatered sludge produced the least GHG emissions. Although dried sludge co-incineration can reduce GHG emissions from transportation and incineration, and electricity generation during the incineration process can offset GHG emissions. However, the drying process produced a large amount of GHG emissions, which still caused large GHG emissions overall.

5.2 Future perspectives

In this study, the standard composition of sewage sludge in Japan, the effect of co-incineration of sewage sludge and MSW, and the potential of co-incineration in Taiwan were clarified. For future research prospects, we propose the following:

(1) It was mentioned in Chapter 3 that metal phosphides are more stable than metal oxides, indicating that the chemical status of heavy metal has a great influence on its characteristics. To properly recycle the sewage sludge and reduce the negative impact of recycling, the element concentration of heavy metals was investigated in this study, but the chemical status of heavy metals was not investigated. Therefore, it is worthy of further investing.

(2) The co-incineration experiment in this study used dried sewage sludge and MSW and was carried out on a laboratory scale. The operating condition of the sample and incineration furnace space, etc. are different from the actual MSWI. Therefore, it is necessary to compare and verify the result with the actual situation.

(3) In the feasibility assessment of co-incineration in Taiwan, only the GHG emission comparison was conducted for the co-incineration of dewatered or dried sewage sludge. However, the cost involved is also an important factor that affects the cooperation in actual implementation. Thus, analysis of costs also needs to be discussed in the future.

Appendix

A1 Combustible sulfur and total sulfur

A 1983 survey conducted in Japan indicated that combustible sulfur content comprised 66% of the total sulfur, (Ministry of Construction and Japan Sewage Works Agency, 1984). Recently, a Romanian study reported that combustible sulfur accounted for 88% of total sulfur in 2018 [1]. In this study, five sludge samples were analyzed, varying in total sulfur content. The combustible sulfur content was determined using an adiabatic bomb calorimeter (CA-4J; Shimadzu Co. Ltd.) and evaluated according to the Japan Industrial Standard [2]. The principle of combustible sulfur analysis is to measure the concentration of sulfur in a sample by completely burning the sample and absorbing the gas generated in water to form a sample solution, and then analyzing the sample solution with ion chromatography.

Samples

The sludge samples were from WWTPs 2, 24, 27, 29, 32 in winter, which contained the sludge with the highest and lowest sulfur concentrations.

Experimental method

About 0.5 g of dried sludge was wrapped in gampi-shi and placed in a sample dish attached to a cylinder. After injecting oxygen so that the pressure in the cylinder was 2-3 MPa, the cylinder was placed in the inner tank and the sample was burned. After combustion, the sample dish and inner surface of the lid were thoroughly washed with ion-exchanged water and the whole solution was collected in a 500 mL beaker (solution 1). The sample dish was placed in a 50 mL beaker, water was added until the sample dish was completely immersed, and the sample dish was heated to boiling for 3-4 minutes (solution 2). Two obtained solutions (solutions 1 and 2) were suction-filtered and ion-exchanged water was added to make a volume of 500 mL. The liquid obtained by filtration was measured using ion chromatography: 0.2369 g of $(COOH)_2$ (containing 95% oxalic acid; Nacalai Tesque) was added to 1 L of ultrapure water; 0.0687 g of Na_2CO_3 (Nacalai Tesque) and 1.013 g $NaHCO_3$ (Nacalai Tesque) were added to a 1 L volumetric flask, which was used as the mobile phase. As a standard reagent for preparing a calibration curve (100%, 50%, 25%, 10%, 5%, 1%, 0.5%), Anion Mixed Standard Solution IV (F^-: 5 mg/L, Cl^-: 10 mg/L, NO_2^-: 15 mg/L, Br^-: 10 mg/L, NO_3^-: 30 mg/L, PO_4^{3-}: 30 mg/L, SO_4^{2-}: 40 mg/L; Kanto Chemical Co., Inc., Tokyo, Japan) was used for dilution with ultrapure water. Each sample was diluted 10 times and then placed in a vial, and the concentrations of SO_4^{2-} were determined using the ion chromatograph (IC) (HIC-20ASUPER, Shimadzu). SO_4^{2-} was converted to S, and the concentration of S in the sample was calculated.

Result

A comparison between total S and combustible S is shown in Table A1.

The combustible S measured by the bomb calorimeter and IC accounted for 104% of the total sulfur on average. The R value was 0.9816 (y = 1.025x; thus, the combustible S and total S can be regarded as equal. The proportion of combustible S to total S was 66% in Japan in 1984, increased to 88% in Romania in 2018, and was nearly 100% in this study. A possible reason for this increase is that there is a large amount of SO_4^{2-} in wastewater due to processes such as acid washing, and advances in technology and knowledge have reduced the inorganic S content of domestic wastewater.

Table A1 Concentrations of total and combustible sulfur.

No.	XRF	IC	Proportion
	T-S (mg/kg DS)	C-S (mg/kg DS)	
27	30,550	32,300	106%
29	6,600	8,330	126%
24	14,800	14,700	99%
2	22,000	21,700	99%
32	13,400	12,200	91%
Average			104%

*XRF: X-ray fluorescence spectrometer, T-S: total sulfur, C-S: combustible sulfur.
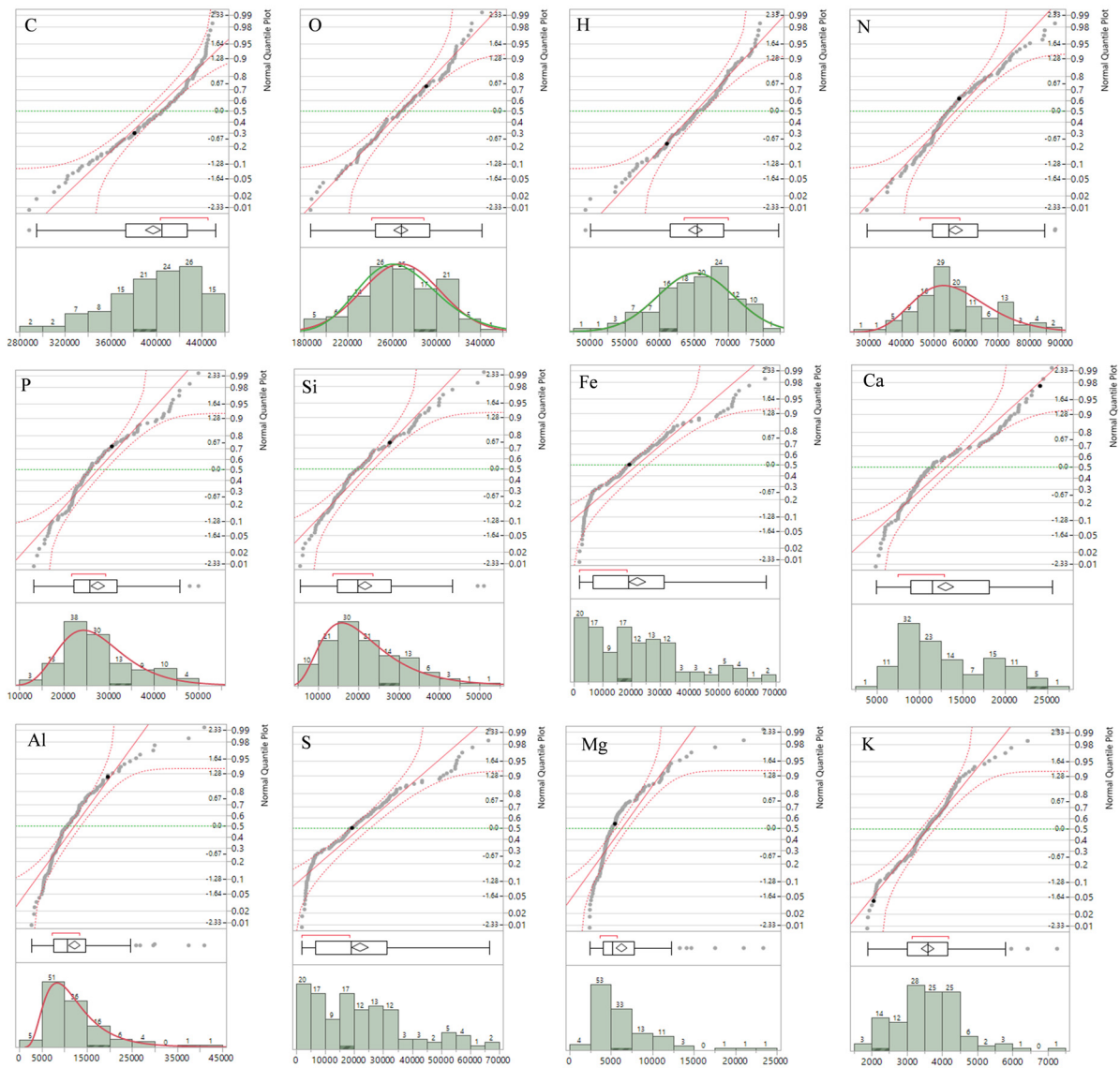
Figure A2.1 Element concentration distribution and verification results for sludge in Japan using JMP software. The gray points in the above figure represent the concentration distribution of the sample (the black point is average), and the red curve represents the 95% confidence interval. The box plot shows the minimum, average, median, maximum, lower quartile (25%), and upper quartile (75%).
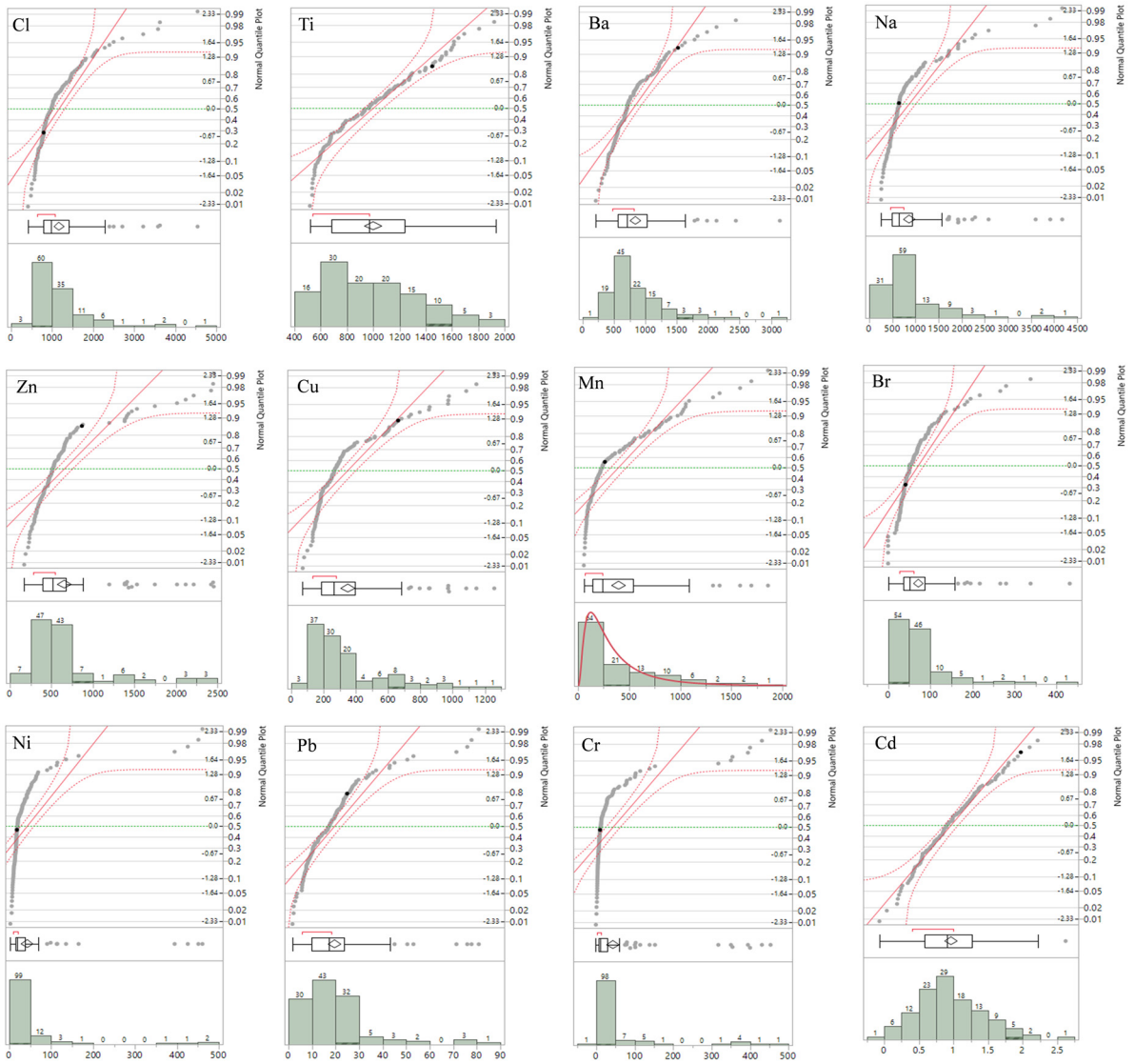
Figure A2.1 cont.
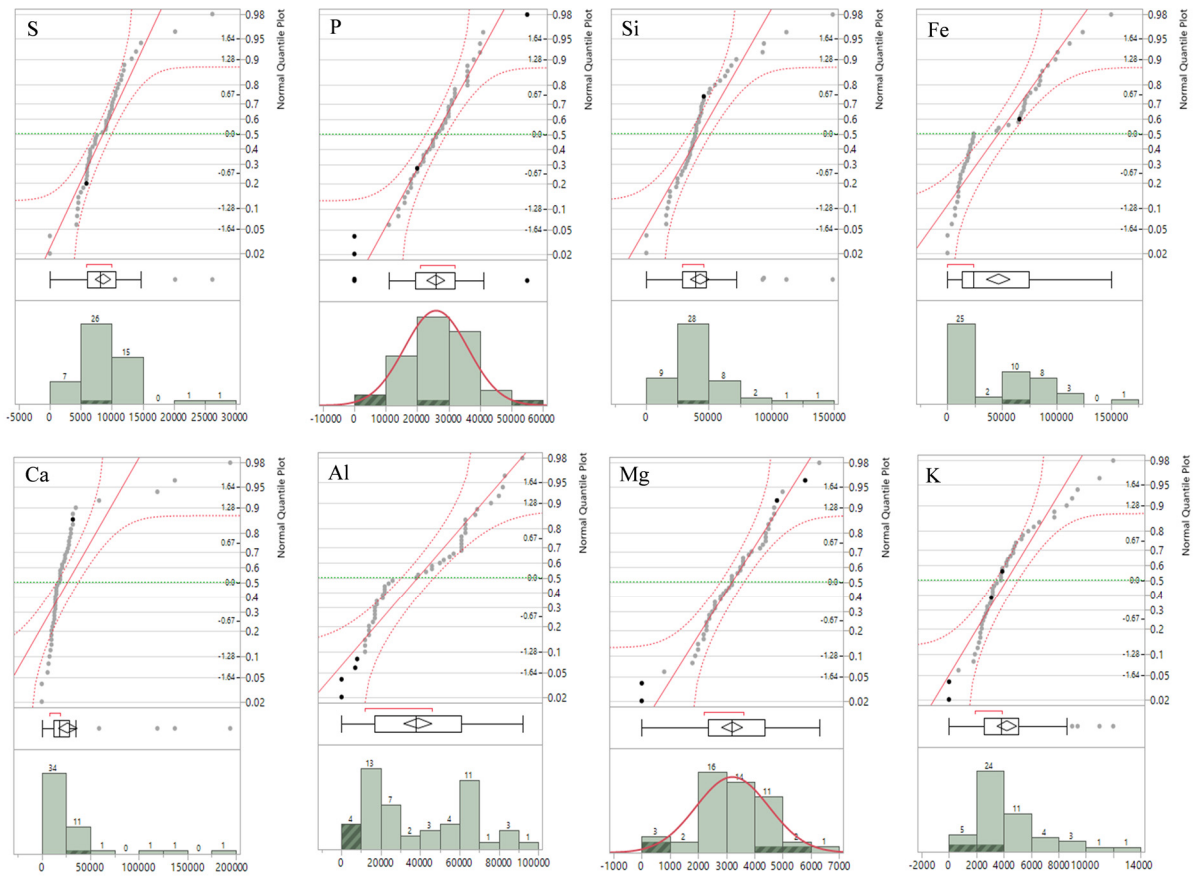
Figure A2.2 Element concentration distribution and verification results for sludge in Sweden using JMP software. The gray points in the above figure represent the concentration distribution of the sample (the black point is average), and the red curve represents the 95% confidence interval. The box plot shows the minimum, average, median, maximum, lower quartile (25%), and upper quartile (75%).
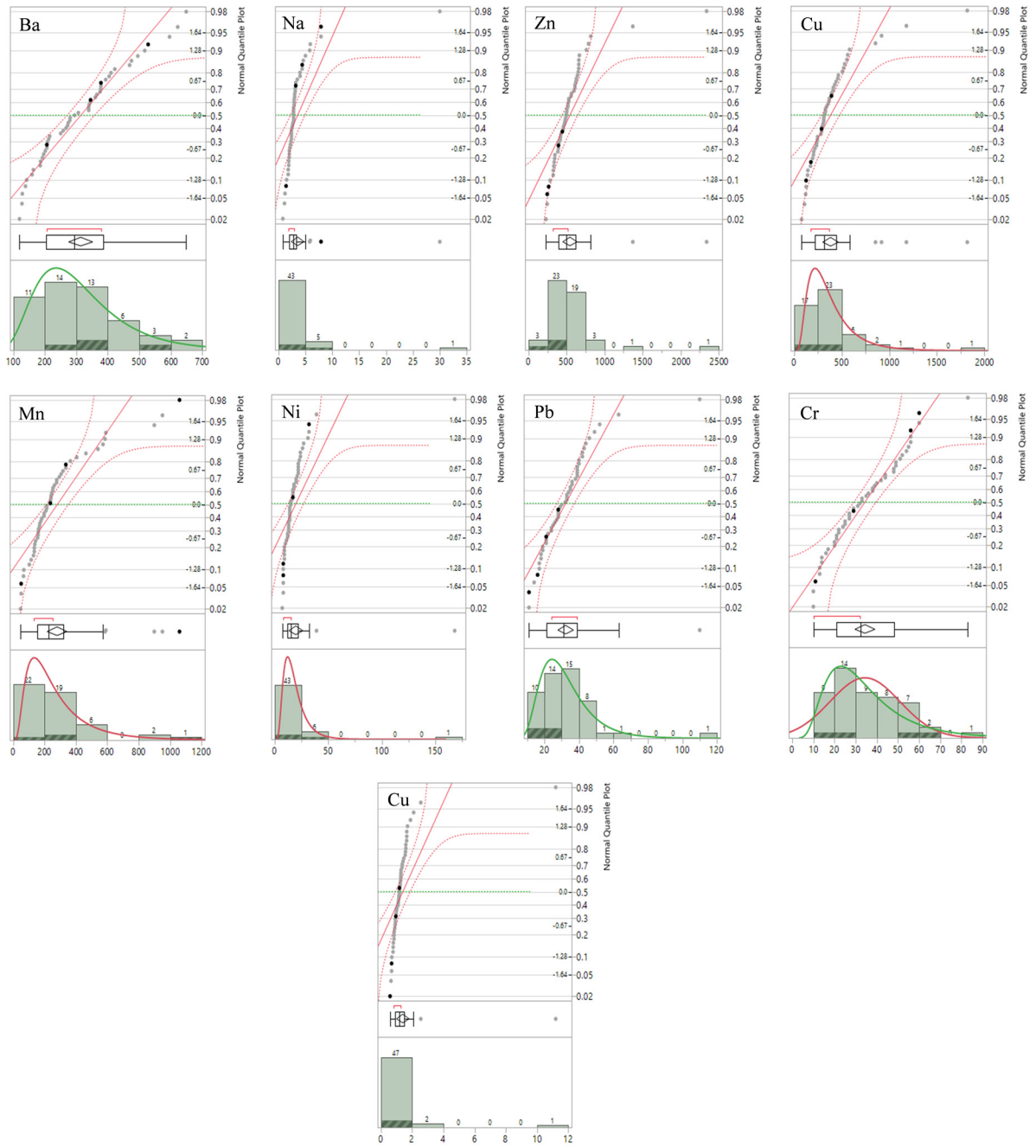
Figure A2.2 cont.
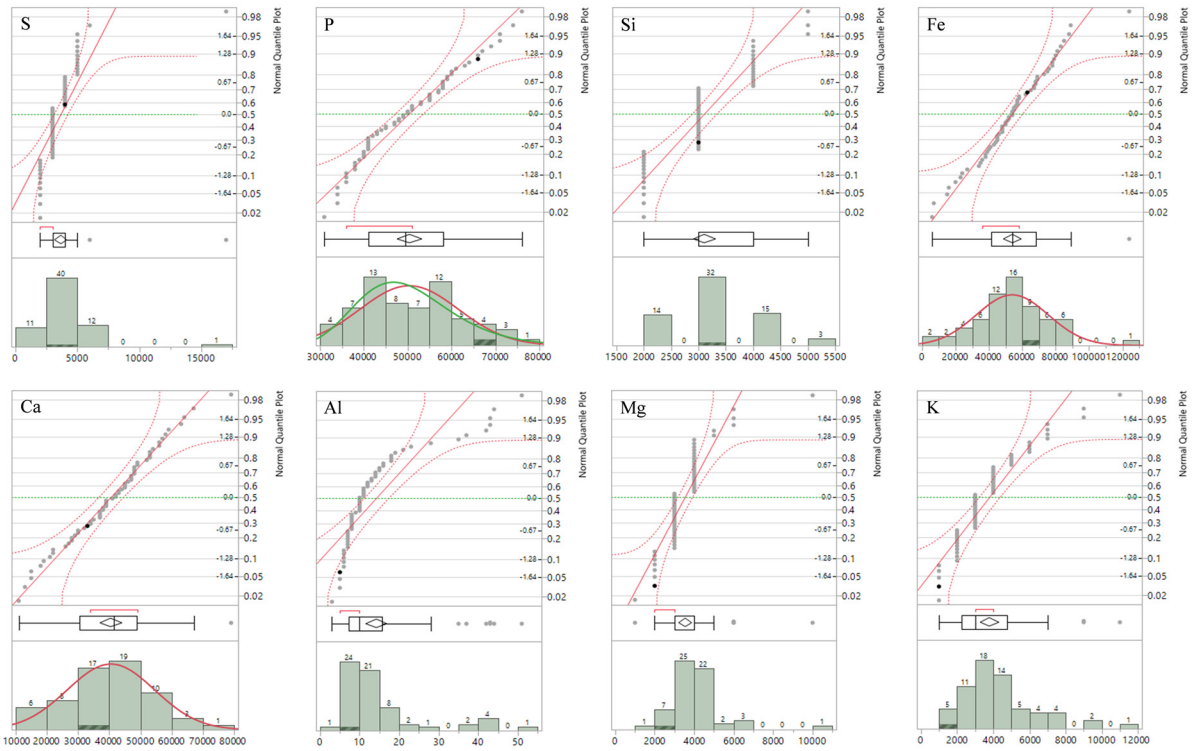
Figure A2.3 Element concentration distribution and verification results for sludge in Switzerland using JMP software. The gray points in the above figure represent the concentration distribution of the sample (the black point is average), and the red curve represents the 95% confidence interval. The box plot shows the minimum, average, median, maximum, lower quartile (25%), and upper quartile (75%).
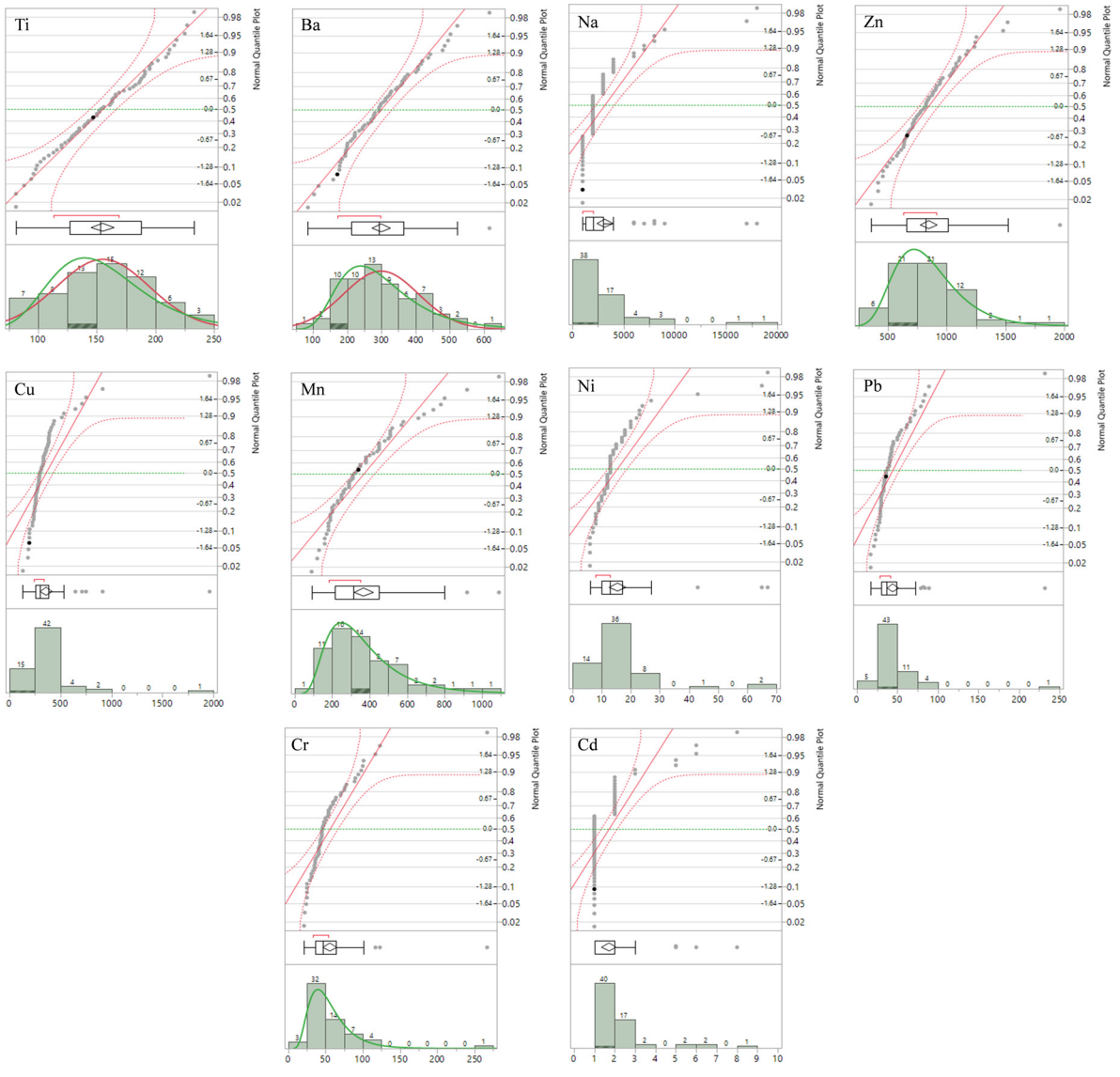
Figure A2.3 cont.
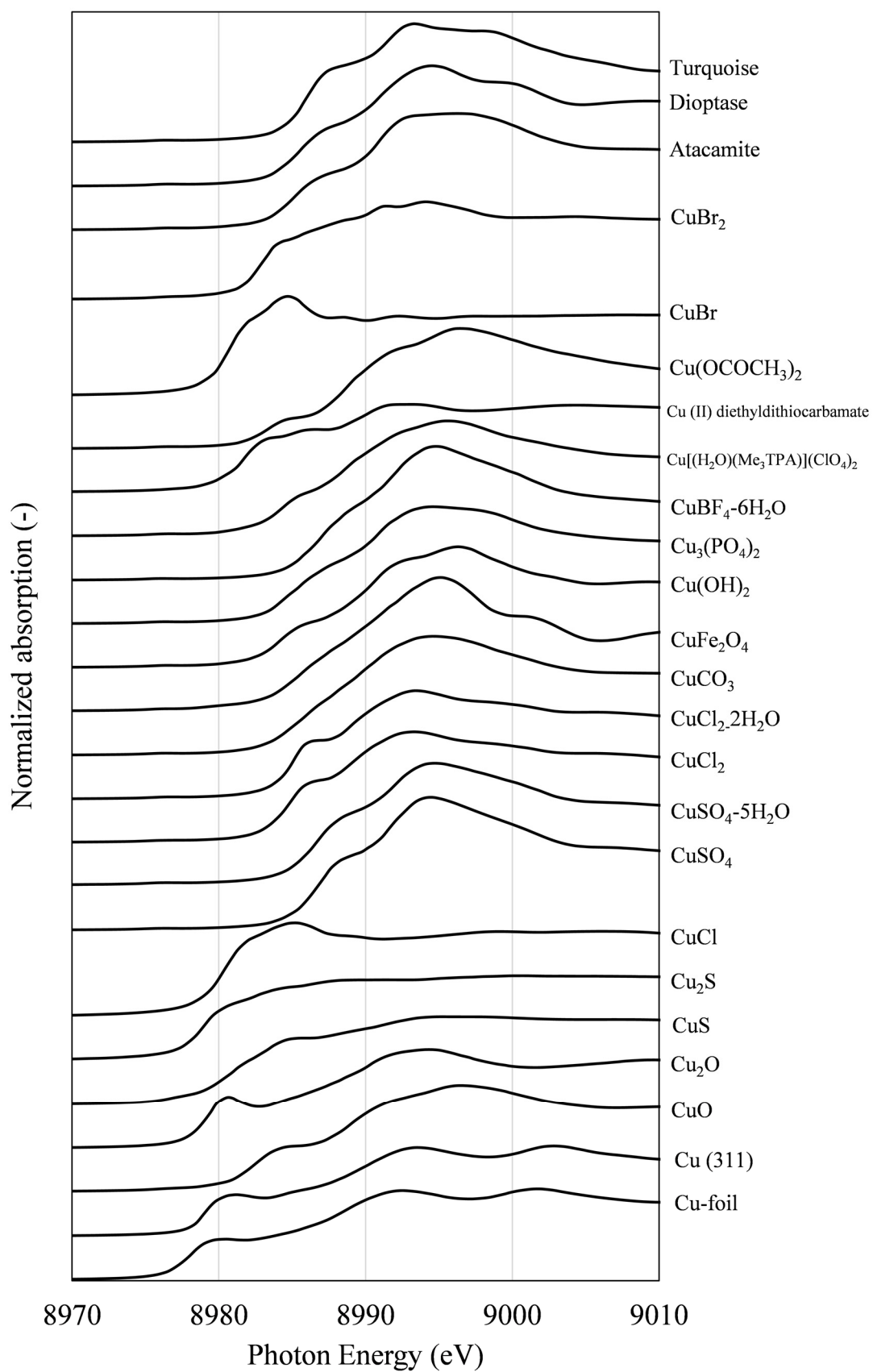
Figure A3 The spectra of Cu K-edge XANES for reference material.

台灣下水污泥與焚化廠連攜評估調查問卷

　　本研究調查目的在於針對污水處理廠及垃圾焚化廠之共同混燒作用。根據先前研究，污泥中富含許多有機質(N, P, S 等化合物)，可與焚化廠操作過程中所產生的重金屬固定化，降低污染發生的可能性。而本研究以全國污水廠與焚化廠資料進行調查，計算各廠之間的距離、混燒率、稼動率及成本計算，進行經濟效益以及環境之評估。

※請以 2017 年之數據回答以下調查問卷內容

@焚化處理場名稱：(　　　　　　　　　　　　　　　)

@焚化爐形式？

　□混燒式機械焚化爐　□全連續機械式爐床　□逆推混燒式機械爐床　□流化床

　□其他（　　　　　　　　　　　　）

@年處理量:(　　　　　　　　　　　　)

@日處理量最高值：(　　　　　　　　) 最低值：(　　　　　　　　　　)

@年售電量:(　　　　　　　　　　)售電價:(　　　　　　　　)

@焚化底灰出廠量與最終處理方式

　　　　灰渣出廠總量(　　　　　　　)

　□掩埋 (　　　　　　)% 　處理單位(　　　　　　　　　　)

　□再利用 (　　　　　　)% 　處理單位(　　　　　　　　)

　□場內佇存 (　　　　　)% 　處理單位(　　　　　　　　　)

　□其他 (　　　　　　　)% 　處理方式、單位(　　　　　　　)

　Ex.灰渣出廠總量(100000t/year)

　　　■掩埋 (80)% 處理單位 (台北市衛生掩埋場)

　　　■再利用(20)% 處理單位(力優勢股份有限公司)

I·【是否有收受污泥】

□否　□有，請繼續往下回答

□下水污泥

　　污泥收受量　（　　　　　　　　）□t/年　□m^3/年

　　污泥含水率　（　　　　　　）％

　　污泥燒失量（　　　　　　）％

　　污泥發熱量　高位（　　　　　　）□cal/kg　□J/kg

　　　　　　　　低位（　　　　　　）□cal/kg　□J/kg

　　污泥收受頻率　□每日　　□（　　　）回/週　　□（　　　）回/月

　　污泥運送方法　□車輛　□配管輸送　□其他（　　　　　　　　）

　　污泥廢棄種類　□一般廢棄　□事業廢棄　□其他（　　　　　　　　）

　　收受來源（污水廠名稱：　　　　　），量（　　　　　　）□t/年　□m^3/年

Figure A4 The example of the questionnaire.

（污水廠名稱： ），量（ ）□t/年 □m³/年

（污水廠名稱： ），量（ ）□t/年 □m³/年

□其他來源（ ）*ex.漁業排水污泥*

　污泥收受量 （ ）□t/年 □m³/年

　污泥含水率 （ ）％

　污泥強熱減量（ ）％

　污泥發熱量高位（ ）□cal/kg □J/kg

　　　　　　低位（ ）□cal/kg □J/kg

　污泥收受頻率 □每日 □（ ）回/週 □（ ）回/月

　污泥運送方法 □車両 □配管輸送 □其他（ ）

　污泥廢棄種類 □一般廢棄 □事業廢棄 □其他（ ）

　污泥收受來源（名稱： ），（ ）□t/年 □m³/年

　　　　　　　（名稱： ），（ ）□t/年 □m³/年

　　　　　　　（名稱： ），（ ）□t/年 □m³/年

Ⅱ‧【收受污泥對焚燒爐的影響】

問 1　污泥、垃圾及其他的混合比率？

　污泥（ ）％ ＋ 垃圾（ ）％ ＋ 其他（ ）％＝合計 100％

　□質量計算 □熱量計算

問 2　在垃圾焚燒和污泥的混合焚燒中，污泥投入焚燒爐的方法有什麼不同嗎？

　□有 □無

　　投入方法的不同（ ）

【投入概要】

垃圾：垃圾⇒垃圾坑⇒垃圾推進器⇒焚燒爐

污泥：污泥⇒污泥接收設施⇒污泥泵⇒焚燒爐

　Ex: ■無，污泥倒入垃圾儲坑照平常模式燃燒

　　　■有，污泥以另外管線配送 or 以污泥泵送入焚燒爐

問 3　在垃圾焚燒和污泥的混合焚燒中，焚化爐操作方法與條件有何不同？

　□有 □無

　　操作方法的不同（ ）

　Ex: ■有，燃燒溫度增加/燃燒時間增加等等具體調整的操作條件

問 4　是否有用於接收污泥和處理的專用設備或設備？「

　□有 □無

　　　名稱（ ）作用（ ）

　　　名稱（ ）作用（ ）

　　　名稱（ ）作用（ ）

　Ex. ■有，名稱（污泥輸送設備）作用（污泥收入、儲存、污泥壓送）

Figure A4 cont.

問5　接收污泥後，燃燒廢氣的性質與收受前有何不同嗎？。

　□有　　□無

　　廢氣性質的不同（　　　　　　　　　　　　　　　　　　　）

　　Ex. ■有，（廢棄酸性增加/廢氣中 S 含量增加…等等）

問6　接收污泥後，焚化底灰渣性質與產生量有何不同？

　性　　質：□有　　□無

　　　　焚化灰渣性質的不同（　　　　　　　　　　　　　　　　　　）

　飛灰產生量：□有　　□無

　　　　產生量的不同（　　　　　　　　　　　　　　　　　）

　　Ex. ■有，底渣中重金屬 Cu,Mg 增加，P 減少

　　　　■有，接收污泥後，底渣的產生量增加 10%　重量

問7　接收污泥後，焚化飛灰渣性質與產生量有何不同？

　性　　質：□有　　□無

　　　　焚化灰渣性質的不同（　　　　　　　　　　　　　　　　　　）

　飛灰產生量：□有　　□無

　　　　產生量的不同（　　　　　　　　　　　　　　　　　）

　　Ex. ■有，飛灰中重金屬增加

　　　　■有，接收污泥後，飛灰的產生量增加 10%　重量

Ⅲ‧【污泥收受歷程】

問8　何時開始收受污泥？

　□（　　　　　）年（　　　　　）月

問9　收受污泥的理由為何？（可重複）

　□當初計畫就有打算收受污泥。

　□幫助污水廠降低污泥處理成本。

　□因鄰近污水廠缺乏污泥處理的能力。

　□因污水廠改變污泥處理方式。

　□由於污水廠污泥處理方法等設施故障。

　□貴設施有足夠且過剩的處理能力。

　□上級組織的政策或指示。

　□行政組織的污泥處理方針的改變。

　□因研究而進行測試。

Figure A4 cont.

□其他

IV・【未來計畫】

問 12　是否有計劃在未來增加或減少驗收數量？

　　□有　　□無

　　□增加　　□減少的理由（　　　　　　　　　　　　　　　　　　）

V・【其它】

問 13　請回答收受污泥的優點與缺點

　　優點

　　缺點

【聯絡方式】

謝謝您的回覆，對於問卷調查有任何的問題及建議，請與聯繫我們。

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Figure A4 cont.

Table A2.1 Combination of scenario 1 which includes the name of facilities, actual distance, mixing ratio, and operating ratio of WWTP/MSWI); the landfill site closest to the MSWI for final disposal of ash.

Combination	WWTP		MSWI		Analysis			Landfill site	
	No	Name	No	Name	Actual distance (km)	Mixing ratio (%)	Operating ratio (%)	Name	Distance (km)
Dewatering sludge									
1	3	Sanyin	8	Shuline	4.4	0.02	56.2	Huiji	13.8
2	15	Zhubei	10	Hsinchu	5.58	0.49	72.5	Hsinchu	5.7
3	1	Dihua	3	Beitou	5.62	1.75	73.3	Bali	19.5
4	58	Fengshanxi	23	Southern region	6.72	1.18	64.2	Daliao	8.3
5	23	Keya	10	Hsinchu	6.78	1.13	73	Hsinchu	5.7
6	54	Gangshan Ciaotou	20	Gangshan	7.69	0.02	73.7	Luzhu	12.5
7	44	Chiayi minhaun	17	Chiayi	11.2	0.36	66.8	Minxiong	24.5
8	50	Annan	18	Chengxi	13.3	0.12	61.9	Chengxi	2.1
9	28	Taichung port	13	Wenshan	13.6	2.3	67	Wenshan	0.3
10	29	Futian	13	Wenshan	13.7	1.23	66.3	Wenshan	0.3
11	56	Zhongqu	23	Southern region	14.1	0.98	64.1	Daliao	8.3
12	55	Nanzi	20	Gangshan	14.2	0.91	74.3	Luzhu	12.5
13	5	Bali	3	Beitou	14.6	1.95	73.4	Bali	19.5
14	59	Dashu	22	Central	15	0.22	64.1	Luzhu	12.8
15	16	Dashi	8	Shuline	15.5	0.13	56.3	Huiji	13.8
16	31	TC harbor	13	Wenshan	16.5	0.04	65.5	Wenshan	0.3
17	4	Danshui	3	Beitou	16.7	0.78	72.6	Bali	19.5
18	61	Liukuaicuo	22	Central region	18.1	1.05	64.6	Luzhu	12.8
19	53	Anping	18	Chengxi	18.2	1.06	62.5	Chengxi	2.1
20	10	Liudu	4	Neihu	18.3	0.17	46.4	Shanzhuku	6.2
21	17	Guishan	8	Shuline	18.6	0.47	56.5	Huiji	13.8
22	6	Linkou	3	Beitou	18.8	0.44	72.3	Bali	19.5
23	45	Chiayi-large	17	Chiayi	21.3	0.2	66.7	Minxiong	24.5
24	9	Pinglin	4	Neihu	24.1	0.003	46.3	Shanzhuku	6.2
25	18	Taoyuan north	8	Shuline	24.2	1.18	56.9	Huiji	13.8
26	48	Liuying	17	Chiayi	24.5	0.12	66.7	Minxiong	24.5
27	14	Zhudong	10	Hsinchu	25.1	0.42	72.4	Hsinchu	5.7
28	51	Huweiliao	20	Gangshan	25.5	0.02	73.7	Luzhu	12.5
29	11	Heping island	4	Neihu	26	0.05	46.3	Shanzhuku	6.2
30	27	Toufen	10	Hsinchu	26.1	0.14	72.2	Hsinchu	5.7
31	46	Puzih	17	Chiayi	27.2	0.17	66.7	Minxiong	24.5
32	35	Buzi	13	Wenshan	28	0.01	65.5	Wenshan	0.3
33	21	Fuhsing	8	Shuline	28	0.001	56.2	Huiji	13.8
34	34	Xinguan	13	Wenshan	28.5	0.01	65.5	Wenshan	0.3
35	19	Shihmen	8	Shuline	29.3	0.1	56.3	Huiji	13.8

Table A2.2 Combination of scenario 2 which includes the name of facilities, actual distance, mixing ratio, and operating ratio of WWTP/MSWI); the landfill site closest to the MSWI for final disposal of ash.

Combination	WWTP		MSWI		Analysis			Landfill site	
	No	Name	No	Name	Actual distance (km)	Mixing ratio (%)	Operating ratio (%)	Name	Distance (km)
Dried sludge									
1	1	Dihua	3	Beitou	5.62	0.41	72.3	Bali	19.5
2	55	Nanzi	20	Gangshan	14.21	0.04	73.7	Luzhu	12.5
3	4	Danshui	3	Beitou	16.7	0.11	72.1	Bali	19.5
4	53	Anping	18	Chengxi	18.2	0.02	61.8	Chengxi	2.1
5	45	Chiayi-large	17	Chiayi	21.3	0.02	66.6	Minxiong	24.5
6	14	Zhudong	10	Hsinchu	25.1	0.09	72.2	Hsinchu	5.7
7	27	Toufen	10	Hsinchu	26.13	0.0004	72.1	Hsinchu	5.7
Dewatering sludge									
1	3	Sanyin	8	Shuline	4.4	0.02	56.2	Huiji	13.8
2	15	Zhubei	10	Hsinchu	5.58	0.49	72.5	Hsinchu	5.7
3	58	Fengshanxi	23	Southern region	6.72	1.18	64.2	Daliao	8.3
4	23	Keya	10	Hsinchu	6.78	1.13	73	Hsinchu	5.7
5	54	Gangshan Ciaotou	20	Gangshan	7.69	0.02	73.7	Luzhu	12.5
6	44	Chiayi minhaun	17	Chiayi	11.15	0.36	66.8	Minxiong	24.5
7	50	Annan	18	Chengxi	13.3	0.12	61.9	Chengxi	2.1
8	28	Taichung port	13	Wenshan	13.6	2.3	67	Wenshan	0.3
9	29	Futian	13	Wenshan	13.7	1.23	66.3	Wenshan	0.3
10	56	Zhongqu	23	Southern region	14.1	0.98	64.1	Daliao	8.3
11	55	Nanzi	20	Gangshan	14.2	0.77	74.2	Luzhu	12.5
12	5	Bali	3	Beitou	14.6	1.95	73.4	Bali	19.5
13	59	Dashu	22	Central region	15	0.22	64.1	Luzhu	12.8
14	16	Dashi	8	Shuline	15.5	0.13	56.3	Huiji	13.8
15	31	TC harbor	13	Wenshan	16.5	0.04	65.5	Wenshan	0.3
16	4	Danshui	3	Beitou	16.7	0.46	72.3	Bali	19.5
17	61	Liukuaicuo	22	Central region	18.1	1.05	64.6	Luzhu	12.8
18	53	Anping	18	Chengxi	18.2	1	62.4	Chengxi	2.1
19	10	Liudu	4	Neihu	18.3	0.17	46.4	Shanzhuku	6.2
20	17	Guishan	8	Shuline	18.6	0.47	56.5	Huiji	13.8
21	6	Linkou	3	Beitou	18.8	0.44	72.3	Bali	19.5
22	45	Chiayi-large	17	Chiayi	21.3	0.15	66.7	Minxiong	24.5
23	9	Pinglin	4	Neihu	24.1	0.003	46.3	Shanzhuku	6.2
24	18	Taoyuan north	8	Shuline	24.2	1.18	56.9	Huiji	13.8
25	48	Liuying	17	Chiayi	24.5	0.12	66.7	Minxiong	24.5
26	14	Zhudong	10	Hsinchu	25.1	0.08	72.2	Hsinchu	5.7
27	51	Huweiliao	20	Gangshan	25.5	0.02	73.7	Luzhu	12.5
28	11	Heping island	4	Neihu	26	0.05	46.3	Shanzhuku	6.2
29	27	Toufen	10	Hsinchu	26.1	0.14	72.2	Hsinchu	5.7
30	46	Puzih	17	Chiayi	27.2	0.17	66.7	Minxiong	24.5
31	35	Buzi	13	Wenshan	28	0.01	65.5	Wenshan	0.3
32	21	Fuhsing	8	Shuline	28	0.001	56.2	Huiji	13.8
33	34	Xinguan	13	Wenshan	28.5	0.01	65.5	Wenshan	0.3
34	19	Shihmen	8	Shuline	29.3	0.1	56.3	Huiji	13.8

Table A2.3 Combination of scenario 3 which includes the name of facilities, actual distance, mixing ratio, and operating ratio of WWTP/MSWI); the landfill site closest to the MSWI for final disposal of ash.

Combination	WWTP		MSWI		Analysis			Landfill site	
	No	Name	No	Name	Actual distance (km)	Mixing ratio (%)	Operating ratio (%)	Name	Distance (km)
Dried sludge									
1	1	Dihua	3	Beitou	5.62	0.41	72.3	Bali	19.5
2	58	Fengshanxi	23	Southern region	6.72	0.71	63.9	Daliao	8.3
3	23	Keya	10	Hsinchu	6.78	0.33	72.4	Hsinchu	5.7
4	29	Futian	13	Wenshan	13.7	0.44	65.7	Wenshan	0.3
5	55	Nanzi	20	Gangshan	14.2	0.28	73.9	Luzhu	12.5
6	5	Bali	3	Beitou	14.6	0.8	72.6	Bali	19.5
7	4	Danshui	3	Beitou	16.7	0.25	72.2	Bali	19.5
8	61	Liukuaicuo	22	Central region	18.1	0.31	64.1	Luzhu	12.8
9	53	Anping	18	Chengxi	18.2	0.33	62	Chengxi	2.1
10	17	Guishan	8	Shuline	18.6	0.14	56.3	Huiji	13.8
11	6	Linkou	3	Beitou	18.8	0.14	72.1	Bali	19.5
12	45	Chiayi-large	17	Chiayi	21.3	0.06	66.6	Minxiong	24.5
13	18	Taoyuan north	8	Shuline	24.2	0.3	56.4	Huiji	13.8
14	14	Zhudong	10	Hsinchu	25.1	0.11	72.2	Hsinchu	5.7
15	27	Toufen	10	Hsinchu	26.1	0.04	72.2	Hsinchu	5.7
16	46	Puzih	17	Chiayi	27.2	0.05	66.6	Minxiong	24.5
Dewatering sludge									
1	3	Sanyin	8	Shuline	4.4	0.02	56.2	Huiji	13.8
2	15	Zhubei	10	Hsinchu	5.58	0.49	72.5	Hsinchu	5.7
3	54	Gangshan Ciaotou	20	Gangshan	7.69	0.02	73.7	Luzhu	12.5
4	44	Chiayi minhaun	17	Chiayi	11.2	0.36	66.8	Minxiong	24.5
5	50	Annan	18	Chengxi	13.3	0.12	61.9	Chengxi	2.1
6	28	Taichung port	13	Wenshan	13.6	2.3	67	Wenshan	0.3
7	56	Zhongqu	23	Southern region	14.1	0.98	64.1	Daliao	8.3
8	59	Dashu	22	Central region	15	0.22	64.1	Luzhu	12.8
9	16	Dashi	8	Shuline	15.5	0.13	56.3	Huiji	13.8
10	31	TC harbor	13	Wenshan	16.5	0.04	65.5	Wenshan	0.3
11	10	Liudu	4	Neihu	18.3	0.17	46.4	Shanzhuku	6.2
12	9	Pinglin	4	Neihu	24.1	0	46.3	Shanzhuku	6.2
13	48	Liuying	17	Chiayi	24.5	0.12	66.7	Minxiong	24.5
14	51	Huweiliao	20	Gangshan	25.5	0.02	73.7	Luzhu	12.5
15	11	Heping island	4	Neihu	26	0.05	46.3	Shanzhuku	6.2
16	35	Buzi	13	Wenshan	28	0.01	65.5	Wenshan	0.3
17	21	Fuhsing	8	Shuline	28	0.001	56.2	Huiji	13.8
18	34	Xinguan	13	Wenshan	28.5	0.01	65.5	Wenshan	0.3
19	19	Shihmen	8	Shuline	29.3	0.1	56.3	Huiji	13.8

Table A2.4 Combination of scenario 4 which includes the name of facilities, actual distance, mixing ratio, and operating ratio of WWTP/MSWI); the landfill site closest to the MSWI for final disposal of ash.

Combination	WWTP		MSWI		Analysis			Landfill site	
	No	Name	No	Name	Actual distance (km)	Mixing ratio (%)	Operating ratio (%)	Name	Distance (km)
Dried sludge									
1	3	Sanyin	8	Shuline	4.4	0.02	56.2	Huiji	13.8
2	15	Zhubei	10	Hsinchu	5.58	0.17	72.3	Hsinchu	5.7
3	1	Dihua	3	Beitou	5.62	0.41	72.3	Bali	19.5
4	58	Fengshanxi	23	Southern region	6.72	0.74	63.9	Daliao	8.3
5	23	Keya	10	Hsinchu	6.78	0.33	72.4	Hsinchu	5.7
6	54	Gangshan Ciaotou	20	Gangshan	7.69	0.03	73.7	Luzhu	12.5
7	44	Chiayi minhaun	17	Chiayi	11.2	0.11	66.6	Minxiong	24.5
8	50	Annan	18	Chengxi	13.3	0.06	61.8	Chengxi	2.1
9	28	Taichung port	13	Wenshan	13.6	0.87	66	Wenshan	0.3
10	29	Futian	13	Wenshan	13.7	0.45	65.7	Wenshan	0.3
11	56	Zhongqu	23	Southern region	14.1	0.45	63.7	Daliao	8.3
12	55	Nanzi	20	Gangshan	14.2	0.28	73.9	Luzhu	12.5
13	5	Bali	3	Beitou	14.6	0.81	72.6	Bali	19.5
14	59	Dashu	22	Central region	15	0.06	64	Luzhu	12.8
15	16	Dashi	8	Shuline	15.5	0.04	56.2	Huiji	13.8
16	31	TC harbor	13	Wenshan	16.5	0.05	65.5	Wenshan	0.3
17	4	Danshui?	3	Beitou	16.8	0.26	72.2	Bali	19.5
18	61	Liukuaicuo	22	Central region	18.1	0.27	64.1	Luzhu	12.8
19	53	Anping	18	Chengxi	18.2	0.34	62	Chengxi	2.1
20	10	Liudu	4	Neihu	18.3	0.06	46.3	Shanzhuku	6.2
21	17	Guishan	8	Shuline	18.6	0.14	56.3	Huiji	13.8
22	6	Linkou	3	Beitou	18.8	0.14	72.1	Bali	19.5
23	45	Chiayi-large	17	Chiayi	21.3	0.06	66.6	Minxiong	24.5
24	9	Pinglin	4	Neihu	24.1	0	46.3	Shanzhuku	6.2
25	18	Taoyuan north	8	Shuline	24.2	0.31	56.4	Huiji	13.8
26	48	Liuying	17	Chiayi	24.5	0.04	66.6	Minxiong	24.5
27	14	Zhudong	10	Hsinchu	25.1	0.11	72.2	Hsinchu	5.7
28	51	Huweiliao	20	Gangshan	25.5	0	73.7	Luzhu	12.5
29	11	Heping island	4	Neihu	26	0.02	46.3	Shanzhuku	6.2
30	27	Toufen	10	Hsinchu	26.1	0.04	72.2	Hsinchu	5.7
31	46	Puzih	17	Chiayi	27.2	0.05	66.6	Minxiong	24.5
32	35	Buzi	13	Wenshan	28	0.002	65.5	Wenshan	0.3
33	21	Fuhsing	8	Shuline	28	0.0002	56.2	Huiji	13.8
34	34	Xinguan	13	Wenshan	28.5	0.02	65.5	Wenshan	0.3
35	19	Shihmen	8	Shuline	29.3	0.03	56.2	Huiji	13.8

Table A2.5 Combination of scenario 0 which includes the name of facilities, and the distance of landfill site closest to the WWTP for final disposal of ash.

Combination	No	WWTP	Landfill site	Distance (km)
1	1	Dihua	Shanzhuku	16.1
2	4	Danshui	Bali	23.7
3	5	Bali	Bali	6.0
4	6	Linkou	Bali	11.4
5	9	Pinglin	Shanzhuku	19
6	10	Liudu	Shanzhuku	13.1
7	11	Heping island	Shanzhuku	25.1
8	3	Sanyin	Huiji	12.5
9	16	Dashi	Dianzihu	19.9
10	17	Guishan	Huiji	7.5
11	18	Taoyuan north	Zhongfu	8.8
12	19	Shihmen	Zhongfu	35.5
13	21	Fuhsing	Zhongfu	40.8
14	14	Zhudong	Emei	15.8
15	15	Zhubei	Xinfeng	9.0
16	23	Keya	Jinshui	1.4
17	27	Toufen	Toufen	2.7
18	28	Taichung port	Wenshan	12.6
19	29	Futian	Dali	8.7
20	31	TC harbor	Hemei	12.9
21	34	Xinguan	Dali District	10
22	35	Buzi	Dali District	10.9
23	44	Chiayi minhaun	Minxiong	11.4
24	45	Chiayi-large	Minxiong	11.4
25	46	Puzih	Xingang	13.6
26	48	Liuying	Yanshui	11.2
27	50	Annan	Anding	11.3
28	53	Anping	Anding	14.7
29	51	Huweiliao	Rende	9.0
30	54	Gangshan Ciaotou	Yanchao	9.9
31	55	Nanzi	Yanchao	16.1
32	59	Dashu	Daliao	12.2
33	61	Liukuaicuo	Daliao	12.5
34	56	Zhongqu	Daliao	16.9
35	58	Fengshanxi	Daliao	5.5

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