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RESEARCH ARTICLE

Assessment of popular techniques for co-processing municipal solid waste in Chinese cement kilns

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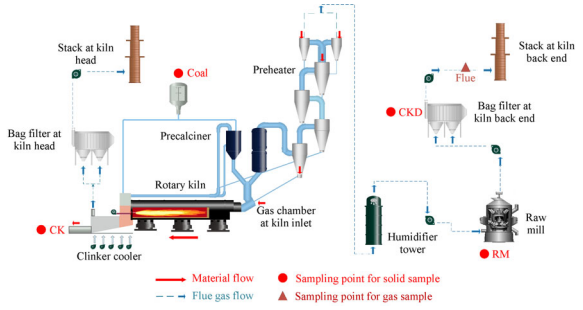
HIGHLIGHTS

- Municipal solid waste (MSW) was fermented, screened, gasified, then co-processed.
- Co-processing MSW in cement kilns could cause excessive pollutant emissions.
- Bypass flue gas can be disposed of through the main flue system.
- Popular MSW co-processing methods do not affect cement quality.

GRAPHIC ABSTRACT



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ABSTRACT

Cement kiln co-processing techniques have been developed in the past 20 years in China, and more than 60 factories now use fermentation, screening, and gasification pre-treatment techniques to co-process municipal solid waste (MSW). There three complete MSW pre-treatment techniques, co-processing procedures, and environmental risk assessments have been described in few publications. In this study, we assessed the effectiveness of each technique. The results suggested that the pollutant content released by each pre-treatment technology was lower than the emission standard. To reveal the mechanisms of pollutant migration and enrichment, the substances in the kiln and kiln products are investigated. The input of co-processing materials (Co-M) produced by fermentation caused formation of polychlorinated dibenzo-p-dioxins and dibenzofuran (PCDD/Fs) in the bypass flue gas (By-gas) in excess of the regulatory standard. The Co-M input produced by the screening and gasifier technologies caused the total organic carbon (TOC) concentration to exceed the standard. In addition, the NO_x, TOC, and PCDD/Fs in the By-gas exceeded the regulatory standard. Raw meal was the primary chlorine and heavy metals input stream, and clinker (CK) and cement kiln dust (CKD) accounted for >90% of the total chlorine output stream. Flue gas and CKD were the primary volatile heavy metal (Hg) output streams. Greater than 70% of the semi-volatile heavy metals (Cd, Pb, Tl and Se) distributed in hot raw meal and bypass cement kiln dust. The low-volatility heavy metals were concentrated in the CK. These results indicated that co-processing techniques used in China still require improvement.

1 Introduction

It has been estimated that 11.20% of all the municipal solid waste (MSW) produced worldwide is produced in China, which has a large percentage (18.41%) of the world population (Mian et al., 2017). The China Statistical Yearbook states that 242.06 million tonnes of MSW were produced in China in 2019 (Fig. S1) (National Bureau of Statistics of China, 2019). The amount of MSW produced in China is expected to increase during the next few decades (Xiao and Zhou, 2020).

Landfill and incineration are currently the primary MSW disposal methods (Yao et al., 2019). However, these methods have some limitations. Rapidly increasing global population and rising sea levels have decreased the amount of land available for landfills, which has indirectly

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increased landfill costs (Ferronato et al., 2019). MSW is not completely destroyed in landfill sites, and leaks cause long-term secondary environmental pollution problems (Sun et al., 2019). Additionally, MSW contains many potentially valuable secondary resources (Cimpan et al., 2015), and incinerating MSW wastes these resources.

The concept of co-processing originated from the concept of the Zero Waste Alliance. Co-processing is the use of waste as a source of energy, or as raw material, in an effort to replace natural mineral resources and fossil fuels in industrial processes, primarily in energy intensive industries such as the generation industries of cement, lime, steel, glass, and power (Wehenpohl et al., 2006).

There are many types of kilns that can co-process waste, but only co-processing techniques for cement kilns have been widely promoted worldwide (Yang et al., 2019). In the 1970s, cement kiln co-processing techniques became popular in many developed countries (Lamas et al., 2013). Research into cement kiln co-processing techniques began in Brazil in the early 1990s, and the relevant national standard was established in 1999 (Lamas et al., 2013). Development of kiln co-processing techniques began in China since 2000. In 2019, 86 cement kilns were licensed to co-process hazardous waste, 63 cement kilns were licensed to co-process municipal sludge, and 60 cement kilns were licensed to co-process MSW in China. Thirty-four cement kilns are currently being built or planned for co-processing MSW in China.

Co-processing MSW in cement kilns has various advantages. Due to many cement kiln plants existing, co-processing techniques require few investments for disposal facility. Meanwhile, there are no effect on cement product quality (Alves, 1993). MSW is completely decomposed as a supplementary fuel or alternative raw material in a cement kiln, which has been regarded as a promising technique for safe destruction of MSW (Conesa et al., 2011). However, heavy metals (HMs) and polychlorinated dibenzo-p-dioxins and dibenzofuran (PCDD/Fs) emissions are problematic when combusting solid waste (Schuhmacher et al., 2009; Liu et al., 2015). Some studies have shown that HMs can be immobilized by solidification and stabilization during the clinker (CK) production process (Zhang et al., 2009; Yan et al., 2018). Appropriate combustion conditions can ensure that PCDD/Fs decompose to yield small molecules of carbon and hydrogen (Xiao et al., 2018). Moreover, refuse-derived fuel (RDF) prepared from MSW decreases both cement production costs and CO_2 emissions (Kara, 2012).

Hasanbeigi et al. (2010) compared the different amounts of energy used in 16 cement kilns in Shandong Province (China) and the international best practices. Unfortunately, the study included plants in only a few regions, and the risks exposed to the environment by the cement production processes were not assessed. Furthermore, many studies have not fully reported the environmental risks during pre-treatment processes and the CK production process (Yang et al., 2012; Yan et al., 2014; Jin et al., 2018; Yan et al., 2018; Xiao et al., 2020).

According to our knowledge, the data for MSW co-processing in cement kilns in China indicated that three pre-treatment techniques (fermentation, screening, and gasification) are commonly used. However, the exposure risks of HMs, PCDD/Fs, and other common pollutants to the environment from using these co-processing techniques have not yet been fully assessed in previous reports. Hence, in the present study, we comprehensively assess the exposure risks to the environment by commonly used MSW pre-treatment processes and the CK production process in China.

In this paper, studies were conducted at three representative cement factories of China. Fermentation pre-treatment was studied at a cement plant in Hubei Province (plant A). Screening pre-treatment was studied at a cement plant in Jiangsu Province (plant B). Gasification pre-treatment was studied at a cement plant in Anhui Province (Plant C). The exposure risks to the environment by emissions from the plants were assessed. The chlorine mass balances and HMs circulating ratios were calculated for coal and co-processing materials (Co-M), including MSW or RDF plus inert components (IC), and combustible gas (CG), raw meal (RM), CK, flue gas, bypass flue gas (By-gas), cement kiln dust (CKD), and bypass cement kiln dust (By-CKD). The effectiveness of the bypass system in plant A was tested (labeled A_1 when the bypass system was not used and A_2 when the bypass system was used). The results of this study can provide important reference data for optimization design of cement kiln co-processing procedures.

2 Materials and methods

2.1 Sites

Each plant of A, B, and C utilizes a new dry cement production process involving a five-stage cyclone pre-heater, a precalciner, and a rotary kiln. The bypass system was not opened in tests A_1-Ba, A_1-Co, B-Ba and C-Ba during CK production (where Ba and Co signify the plant baseline test and the co-processed MSW test, respectively). The bypass system was used at the kiln ends in tests A_2-Co, B-Co, and C-Co, which can reduce the possibility of alkali and blockage by ejecting a certain amount of smoke with the highest concentrations of alkali and sulfur compounds (Zhan et al., 2016). Most importantly, the bypass system can slow down chlorine formation caused by crust blocking (Zhan et al., 2016).

2.2 Pre-treatment processes

The complex composition of MSW and the dangerous components may present mean that MSW can not be fed

directly into a kiln. The standard for pollution control on co-processing of solid waste in cement kiln, released by Standardization Administration of China in 2013, requires that the solid waste to be pre-treated before being fed into a kiln. Three MSW pre-treatment techniques are currently commonly used in China (Fig. 1).

2.2.1 Fermentation (plant A pre-treatment method)

The MSW was broken up and then transferred to an aerobic fermentation tank (containing aerobic microorganisms that can degrade kitchen waste) and left for 15 days. The energy produced during fermentation was used to dry the MSW to decrease its moisture content. The MSW was then mechanically separated into combustible components, IC, and metal components. The combustible components were crushed to obtain the refuse derived fuel (RDF), which was directly transferred to the cement kiln precalciner for disposal. The metal components were magnetically separated for recycling. The IC and combustible components could not be completely separated, limiting their value (pure IC was used to produce the RM). If a large amount of IC was added to the raw mill, the total organic carbon (TOC) concentration in the flue gas would be higher than the regulatory standard (Cai et al., 2015). Therefore, the IC was fed into the flue gas chamber at the kiln end for disposal. The fermentation pre-treatment process robustly produced odors that needed to be removed using a biological purification system. The leachate produced during the pre-treatment process was treated and subsequently discharged to the municipal sewage network.

2.2.2 Screening (plant B pre-treatment method)

MSW was screened to separate different components and then crushed. Residues including fabric, kitchen waste, paper, plastic, and large particles of other materials were disrupted by utilizing air separation, coarse crushing, and magnetic separation, and then they were separated while being vibrated. Combustible materials were processed to obtain the RDF, which was transferred to an RDF storage area. Residues, including ceramics, glass, kitchen waste, plastic debris, slag, and small particles of other materials, were mixed and fed into a density separator. Light components (primarily plastic debris) were separated and processed to obtain the RDF. The remaining components (IC) were mixed with the RM and fed into the raw mill for disposal. The MSW pre-treatment center had a sewage treatment system in which the leachate produced during the pre-treatment processes was treated. Odors were removed by using a biological liquid spray and an activated carbon adsorption process.

2.2.3 Gasification (plant C pre-treatment method)

Plant C had an integrated Conch Kawasaki Kiln (CKK) system that combined MSW gasification and a new dry cement production technique (Sharma et al., 2020). MSW was homogenized and crushed in a storage pit and subsequently transferred to the gasifier for combustion. In the gasifier, the MSW was gasified (converted into fuel or synthesis gas) in the presence of oxidants (Arena, 2012). The gasification system contained flowing medium at a high temperature, and a large amount of heat was released when fluidized sand came into contact with MSW. Some of the heat was consumed by the flowing medium, and the remaining energy was consumed to gasify the MSW to obtain a CG at approximately 500°C. The CG was then transferred to the cement kiln for use as fuel. Aluminum, iron, and other metals were separated by using a magnetic separator from the gasification slag discharged from the bottom of the furnace. The residue components (non-metallic slag and dust, including gasifier dust and CKD) were placed in the cement kiln raw mill for disposal (as an RM substitute). Odors produced in the MSW storage pit were removed by using an air extractor. Separated air components were fed into the gasifier to support combustion. The odor-containing gas was sent to the CK cooler to cool the CK. When cement production was stopped, the gas purification device removed the odor. Leachate produced in the waste storage pit was sent to a sewage storage tank through a sewage filter and subsequently injected into the lower end of the precalciner using a delivery pump. The leachate was then treated at a high temperature in the precalciner.

2.3 Clinker production

Cement is the primary component of concrete and primarily produced from RM, which is a mixture of limestone and clay. Limestone and clay contain large amounts of calcium, silicon, aluminum, and iron oxides (Kara, 2012), which qualifies cement significant compressive strength and other useful physical properties. Cement was produced in plants A, B, and C using fuel consisting of coal and Co-M produced in the MSW pre-treatments. The CK production capacities of plants A, B, and C were 6000, 5000, and 5500 t/d, respectively, and the MSW co-processing capacities of plants A, B, and C were 350, 500, and 300 t/d, respectively.

2.4 Test procedure

Baseline and co-processing conditions were used in the plant A, B, and C tests, as shown in Table 1. In each baseline test, the pre-treatment equipment was closed and no pre-treated material was fed into the kiln. The normal CK production process was performed in the cement kiln.

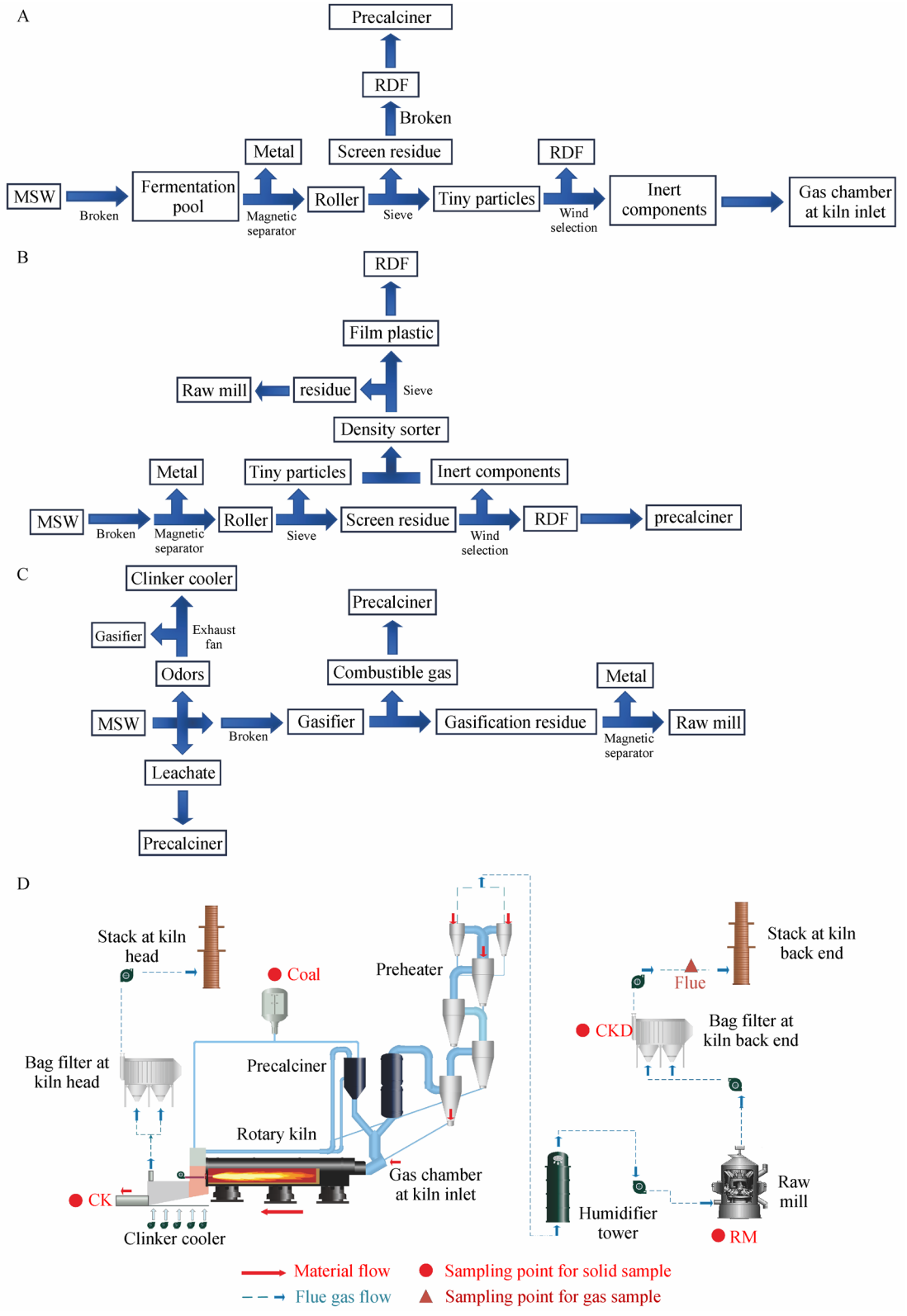

Fig. 1 Pre-treatment processes used in plants A, B, and C. D is the clinker production process.

Table 1 Operating conditions used during baseline and municipal solid waste (MSW) co-processing tests

Project	A_1-Ba	A_1-Co	A_2-Co	B-Ba	B-Co	C-Ba	C-Co
RM feed rate (t/h)	368.08	358.80	319.40	376.50	372.90	365.00	371.00
Coal feed rate (t/h)	33.13	29.27	26.23	29.40	24.91	30.50	31.00
RDF (MSW) feed rate (t/h) [a)]	–	11.54	15.50	–	10.08	–	12.90
CK output rate (t/h)	234.46	231.82	208.76	241.30	240.90	228.20	230.10
Flue gas (Nm^3/h)	4.18×10^5	4.25×10^5	3.23×10^5	4.99×10^5	4.94×10^5	8.04×10^5	7.92×10^5
By-gas (Nm^3/h)	–	–	3.78×10^4	–	1.18×10^4	–	1.91×10^4
CKD output rate (t/h)	18.40	17.94	22.80	18.83	18.65	18.25	18.55
By-CKD output rate (t/h)	–	–	0.52	–	0.38	–	0.10
CK consumption (kWh/t)	31.40	31.34	31.34	62.73	61.59	23.93	24.14
Preprocessing consumption (kWh/t)	–	33.50	33.50	–	14.31	–	32.40
Coal replacement rate (%)	–	10.61	14.9	–	13.22	–	– 1.8
RM replacement rate (%)	–	–	–	–	1.15	–	1.41

Notes: a) Refuse derived fuel (RDF (MSW)) feed rate (t/h), RDF means the material that plants A and B co-processed, MSW means the material that plant C co-processed, By-gas means the bypass flue gas, By-CKD means the bypass cement kiln dust, A_1 means the bypass system was not used, A_2 means the bypass system was used, Ba means the baseline test, and Co means the co-processing test.

In the co-processing tests, plant A was fed with RDF, plant B with RDF and IC, and plant C with MSW, respectively. The RDF feed rates of A_1-Co, A_2-Co, and B-Co were 11.54, 15.50, and 10.08 t/h, respectively. The MSW feed rate of C-Co was 12.9 t/h. The input flow rate, the output flow rate, and the energy consumption of the three plants are shown in Table 1. The odor output rate for the CK cooler in plant C was 1.16×10^4 m^3/h. Three 6-day field experiments were performed to assess the accuracy of the data. Baseline tests were performed for the first 2 days, and co-processing tests were performed for the last 4 days.

2.5 Sample collection and analysis

The flue gas and By-gas samples were collected iso-kinetically from the relevant gas streams. Dust samples were collected from the dust collector, RM from the raw mill, and CK from the conveyor belts located after the CK coolers. Samples were collected from the same sampling points at 30 min intervals and then were analyzed in triplicate. The mean and standard deviation were assessed for each sampling point, and time was allowed to assess the accuracy and validity of the data. All of the tests required the cement plant processing equipment to operate under normal conditions.

The test samples were MSW, odors produced during pre-treatment, IC, coal, RDF, CG, RM, CK, CKD, By-CKD, flue gas, and By-gas. The calorific values and other industrially useful characteristics of the MSW, coal, RDF, CG, RM, and CK samples were determined. The concentrations of various pollutants in the flue gas, By-gas, CKD, and By-CKD samples were determined. The HMs and chlorine concentrations in the input and output samples were also determined. The sampling analyses were performed by using the methods described in current Chinese standard (Table S1).

3 Results and discussion

3.1 Evaluation of the pre-treatment products

The characteristics of the MSW in the three plants are shown Table S2. The results of the proximate analyses of the MSW from the three plants were similar, but the calorific value for the plant A sample was 537.54 kcal/kg lower than that for the plant B sample and 451.5 kcal/kg lower than that for the plant C sample. This may result from the lifestyles of the inhabitants near the different plants. Such differences between the MSW calorific values in different areas are inevitable.

Pre-treatment of MSW using fermentation and screening was utilized to obtain the RDF, which is composed of granular or fluffy combustible solid wastes after removing the IC from the MSW (Kara, 2012). The RDF produced after fermentation accounted for 54% of the MSW, and the RDF produced after screening accounted for 39.2% of the MSW (Fig. S2). The gasification method produced up to 2.27×10^4 m^3/h (Table S3). Table S2 shows that the RDF produced by fermentation and screening had a low calorific value by more than 1000 kcal/kg. The effective component of the CG produced by gasification (Table S3) was more than 80% of the total CG volume. Meanwhile, the coal replacement rates (Table 1) for the A_1-Co, A_2-Co and B-Co were 10.61%, 14.49%, and 13.22%, respectively. Additionally, the RM replacement rates (Table 1) for the B-Co and C-Co were 1.15% and 1.41%, respectively. This indicated that all three pre-treatment methods efficiently

produced the MSW into combustible components.

All of the odors (Table S4) and leachate components were removed effectively, and all of the components produced during the pre-treatment process met the Chinese standards.

3.2 Environmental risk assessments for flue gas and bypass flue gas produced during co-processing

Conesa et al. (2011) found HF concentrations were less than 0.1 mg/m^3 and HCl concentrations range were 0.2–3.7 mg/m^3 in the flue gas emitted from cement kiln stacks. We found HF concentrations were less than 0.1 mg/m^3 and HCl concentrations range were 0.04–0.92 mg/m^3 in flue gas emitted from the stacks of the three cement kiln plants (Table 2), possibly because the contents of chlorine (0.29%–0.58%) and fluorine (0.01%–0.02%) in the MSW were low (Table S2). However, the concentrations of some pollutants in the flue gas emitted during the three co-processing methods were higher than that of the relevant standard.

Table S2 shows that the sulfur concentrations were 0.09%–0.17% in MSW from the three cement kiln plants, which may be the reason for the low SO_2 concentration in the flue gas in the A_1-Ba and A_1-Co tests. However, the SO_2 concentration in the flue gas emitted in the A_2-Co test was slightly higher than the emission limit. This was attributed to a high RDF feed rate (Table 1) that resulted in incomplete RDF combustion.

In plant B, the CO concentrations were higher in the co-processing test samples than that in the baseline test samples, and this caused the TOC concentrations in the co-processing test samples to be 10 times higher than the relevant limit. The carbon concentrations in the MSW were greater than 10% (Table S2), which was caused by the high content of organic matter in kitchen waste. The CO concentrations in the co-processing test samples were high because kitchen waste with a high organic matter content was fed into the raw mill as a substitute for RM. In addition, the RDF volume was affected by the release of CO and TOC during co-processing (Di Lonardo et al., 2016). Large RDF granules could not be suspended during combustion because of their high masses (the size of the RDF in B-Co was approximately 10 times larger than that in A-Co). Hence, RDF fell into the flue gas chamber of the kiln, increasing the CO and TOC emissions.

In plant C, the TOC concentrations in the co-processing test were 8.7 times higher than the relevant limit. This was attributed to incomplete combustion of CG (originating from the MSW) in the precalciner (Tokheim et al., 2001).

The HCl concentration was higher in the A_1-Co flue gas than that in the A_2-Co flue gas. This indicated that the

Table 2 Pollutant concentrations in flue gas and bypass flue gas (O_2 concentration 10%)

Project	Compositions	A_1.Ba	A_1-Co	A_2-Co	B-Ba	B-Co	C-Ba	C-Co	Limit
Flue gas	O_2 (%)	8.10±0.11	8.10±0.12	8.55±0.13	8.90±0.14	8.50±0.15	11.50±0.16	10.90±0.15	–
	CO_2 (%)	23.09±0.40	23.85±0.55	24.50±0.34	22.20±0.08	21.90±0.32	16.80±0.67	18.10±0.75	–
	CO (mg/m^3)	142.00±0.98	238.00±0.91	5230±0.45	279.00±1.21	1964±10.95	36.00±0.40	54.00±0.45	–
	PM (mg/m^3)	0.50±0.01	0.50±0.03	3.90±0.03	31.02±0.55	35.55±0.58	8.40±0.34	23.60±0.28	30
	SO_2 (mg/m^3)	2.60±0.05	2.50±0.07	225.50±0.48	2.72±0.03	2.60±0.03	3.50±0.03	3.30±0.05	200
	NO_x (mg/m^3)	355.00±2.67	358.00±1.02	169.90±2.31	408.00±1.98	331.00±1.45	400.00±0.85	395.00±1.98	400
	NH_3 (mg/m^3)	0.90±0.03	1.20±0.02	1.09±0.01	0.57±0.02	0.62±0.01	0.03±0.01	0.21±0.02	10
	HCl (mg/m^3)	0.12±0.01	0.30±0.02	0.04±0.01	0.67±0.01	0.92±0.03	0.37±0.03	0.22±0.01	10
	HF (mg/m^3)	ND	ND	ND	0.04±0.02	0.05±0.01	0.03±0.01	0.03±0.01	1
	TOC (mg/m^3)	1.74±0.03	2.91±0.02	1.80±0.03	11.5±0.07	118.2±0.98	10.40±0.80	87.00±0.25	10
By-gas	O_2 (%)	–	–	20.60±0.32	–	20.65±0.45	–	19.45±0.03	–
	CO_2 (%)	–	–	0.85±0.02	–	0.65±0.04	–	1.97±0.04	–
	CO (mg/m^3)	–	–	1.50±0.03	–	51.00±0.35	–	95.00±0.25	–
	PM (mg/m$^{3)}$	–	–	10.00±0.15	–	12.70±0.09	–	3.00±0.03	30
	SO_2 (mg/m^3)	–	–	1.50±0.03	–	51.00±0.95	–	21.00±0.11	200
	NO_x (mg/m^3)	–	–	46.50±0.05	–	998.00±2.25	–	1705±2.23	400
	NH_3 (mg/m^3)	–	–	4.33±0.04	–	4.06±0.03	–	0.35±0.04	10
	HCl (mg/m^3)	–	–	0.03±0.01	–	8.72±0.07	–	0.74±0.05	10
	HF (mg/m^3)	–	–	0.05±0.01	–	1.02±0.03	–	0.21±0.03	1
	TOC (mg/m^3)	–	–	2.17±0.10	–	44.34±0.90	–	79.0±0.65	10

Notes: ND means not detected, PM means particulate matter, TOC means total organic carbon, A_1 means that the bypass system was not used. By-gas means the bypass flue gas. A_2 means that the bypass system was used, Ba means the baseline test, and Co means the co-processing test.

bypass system decreased chlorine enrichment in the cement kiln. This was consistent with the findings of Zhan et al. (2016). The NO_x and TOC concentrations in the B-Co and C-Co By-gas samples were higher than that of the relevant standard, because the concentrations of carbon and nitrogen in the MSW were high (Table S2). Additionally, the high O_2 concentration in the By-gas promoted oxidation of nitrogen compounds and organic matter to generate more NO_x (Brown et al., 2014) and TOC (Fan et al., 2014).

Environmental pollution caused by HMs has long been considered an important environmental problem (Clavier et al., 2019; Abd Ali et al., 2020; Yin et al., 2021). It is very important to evaluate HMs emissions in cement kiln flue gas. Table 3 shows that the concentrations of most of the HMs in the flue gas emitted from the three factories were lower than that were found in previous studies (Zemba et al., 2011; Yan et al., 2018). The standard for pollution control on co-processing of solid wastes in cement kiln, released by Standardization Administration of China in 2013, contains flue gas HMs concentration thresholds. These are 50 μg/m^3 for Hg (a volatile HM), 1000 μg/m^3 for Cd, Pb, and Tl (semi-volatile HMs), and 500 μg/m^3 for Co, Cr, Cu, Mn, Ni, Sb, Sn, and V (low-volatility HMs). The HMs concentrations in the flue gas emitted from the plants were all lower than these limits. The HMs concentrations in the By-gas samples were also lower than the limits. The Hg concentrations in flue gas samples from baseline and co-processing tests were very similar in the three plants, which was consistent with a previous study (Kara, 2012). However, the semi-volatile HMs concentrations were slightly lower in the three plants baseline test flue gas (0.32–2.21 μg/m^3) than that in the co-processing test (0.65–3.00 μg/m^3) flue gas, which was consistent with previous findings of Yan et al. (2018), because Co-M (MSW or RDF plus IC) was likely to contribute to a small amount of semi-volatile HMs (Table S5). No significant differences were found between the low-volatile HMs concentrations in the flue gas emitted from the three plants in the baseline (1.20–3.58 μg/m^3) and co-processing tests (1.10–3.00 μg/m^3). The bypass system used in the A_2-Co test slightly increased the Hg and semi-volatile HMs concentrations in the flue gas relative to the concentrations found in the A_1-Co test. This was attributed to the high RDF feed rate. Therefore, feeding the Co-M (MSW or RDF plus IC) did not markedly increase the volatile HM, semi-volatile HMs and low-volatile HMs concentrations.

The risks posed to the environment and to human health by PCDD/Fs released in flue gas emitted from industrial plants are important environmental and public health issues (Schuhmacher et al., 2004; García-Pérez et al., 2015). Xiao et al. (2020) suggested that co-processing less than 3% MSW fly ash in a cement kiln could markedly decrease the risks posed to the environment by PCDD/Fs emitted in flue gas. Yan et al. (2018) identified PCDD/Fs "fingerprints" in samples of the input and output materials of a cement kiln and identified the mechanisms through which PCDD/Fs are formed and destroyed. We assessed the risks exposed by PCDD/Fs emissions in flue gas and By-gas emitted during the three different MSW co-processing procedures. The PCDD/Fs concentrations in the samples from plants A, B, and C were 0.0040–0.0042, 0.439–0.719, and 0.0277–0.0336 ng-TEQ/m^3, respectively (Fig. 2(a)). The limit of standard for pollution control on co-processing of solid wastes in cement kiln, released by Standardization Administration of China in 2013, is 0.1 ng-TEQ/m^3. The PCDD/Fs concentrations in the B-Ba and B-Co samples were clearly higher than the Chinese standard limit. In this study, the PM concentration of B-Ba and B-Co were 31.02 and 35.55 mg/m^3, respectively, and a high PM concentration in the flue gas could cause the PCDD/Fs concentration to be higher than the relevant limit (Karstensen, 2008; Tang et al., 2013). The PCDD/Fs concentrations were lower in the B-Co samples than that in the B-Ba samples, indicating that the plant B co-processing procedure did not increase flue gas PCDD/Fs concentrations. The PCDD/Fs concentrations in the co-processing By-gas samples were higher than the Chinese standard

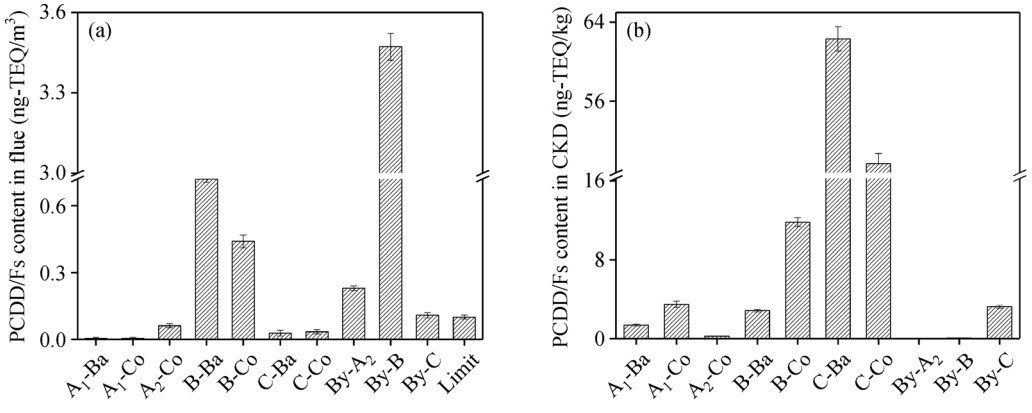

Fig. 2 (a) Polychlorinated dibenzo-*p*-dioxin and dibenzofuran (PCDD/Fs) concentrations in the flue gas and bypass flue gas (By) samples. (b) PCDD/Fs contents of the cement kiln dust (CKD) and bypass cement kiln dust (By) samples. A_1 means that the bypass system was not used. A_2 means that the bypass system was used. Ba means the baseline test. Co means the co-processing test.

Table 3 Heavy metal concentrations in the flue gas and bypass flue gas (By-gas) (O_2 concentration 10%)

Project	Metal (μg/m^3)	A_1-Ba	A_1-Co	A_2-Co	B-Ba	B-Co	C-Ba	C-Co	Zemba et al. (2011)	Yan et al. (2018)
Flue gas	As	0.22±0.04	0.58±0.01	0.10±0.01	0.06±0.01	1.7±0.01	0.12±0.02	0.50±0.03	0.2–4.1	0.17–4.50
	Hg	2.10±0.15	2.10±0.17	49.55±0.32	18±0.19	13±0.14	16±0.15	15±0.29	0.2–27	28.60–61.95
	Cd	ND	0.01	ND	ND	0.19±0.02	ND	0.01±0.01	0.1–37	ND–0.04
	Pb	0.05±0.03	0.02±0.01	0.34±0.02	0.28±0.03	0.07±0.01	2.05±0.02	2±0.05	0.3–88	ND–2.09
	Sn	0.20±0.04	0.10±0.01	0.15±0.02	0.19±0.01	0.04±0.02	0.01±0.01	0.03±0.01	–	–
	Co	0.17±0.01	0.17±0.02	ND	0.18±0.01	0.18±0.02	0.20±0.01	0.22±0.03	0.1–19	0.03–0.18
	Cr	0.34±0.01	0.34±0.04	0.15±0.01	0.50±0.03	0.35±0.01	0.49±0.02	0.43±0.02	0.4–83	6.12–12.38
	Cu	0.17±0.02	0.17±0.01	0.10±0.01	0.18±0.02	0.43±0.01	0.63±0.03	0.80±0.05	0.4–37	0.27–1.16
	Mn	0.17±0.02	0.17±0.03	0.23±0.01	2±0.08	1±0.02	2.12±0.07	2±0.04	0.1–210	2.16–4.50
	Ni	0.03±0.01	0.03±0.02	0.05±0.01	0.03±0.01	0.03±0.01	0.05±0.01	0.03±0.01	0.3–44	0.78–4.23
	Sb	0.07±0.01	0.07±0.02	0.01±0.01	0.07±0.01	0.07±0.01	0.08±0.02	0.09±0.01	0.2–30	0.17–8.75
	Tl	0.04±0.01	0.04±0.01	0.46±0.04	0.04±0.01	0.04±0.01	0.04±0.02	0.05±0.01	0.12–2.8	0.03–1.06
	V	0.03±0.01	0.03±0.01	0.02±0.02	0.04±0.02	0.04±0.01	0.04±0.02	0.04±0.01	0.2–30	0.17–8.75
	Zn	0.58±0.01	0.36±0.09	0.45±0.04	8±0.24	0.26±0.01	8.2±0.09	7±0.08	0.01–329.63	1.30–5.99
	Mo	0.34	0.34	0.01	0.26	0.35	0.32	0.43	–	0.184–0.25
	Be	ND	ND	ND	ND	ND	ND	ND	–	ND
	Tl + Cd + Pb + As	0.32±0.03	0.65±0.03	0.91±0.02	0.38±0.02	2±0.03	2.21±0.04	3±0.01	1000	1.07–5.34
	Be + Cr + Sn + Sb + Cu + Co + Mn + Ni + V	1.20±0.02	1.10±0.04	0.71±0.06	3±0.07	2±0.04	3.58±0.04	3±0.01	500	13.05–22.90
By-gas										
	As			0.10±0.01		1±0.04		6±0.09		
	Hg			2.76±0.04		40±0.41		18±0.21		
	Cd			0.09±0.01		0.05±0.01		3±0.03		
	Pb			6.4±0.05		0.14±0.01		49±0.23		
	Sn			0.15±0.01		1.3±0.03		1±0.12		
	Co			0.02±0.01		3.4±0.04		1±0.05		
	Cr			1.08±0.01		16±0.12		26±0.43		
	Cu			0.37±0.01		3.4±0.06		21±0.05		
	Mn			1.43±0.01		7±0.09		21±0.12		
	Ni			0.42±0.04		0.5±0.01		9±0.12		
	Sb			0.01±0.01		1.4±0.12		0.57±0.01		
	Tl			0.01±0.01		0.8±0.02		0.36±0.01		
	V			0.02±0.01		0.7±0.02		0.28±0.01		
	Zn			0.45±0.08		5.1±0.01		89±0.01		
	Mo			0.01±0.01		6.8±0.12		21±0.09		
	Be			ND		ND		0.02±0.01		
	Tl + Cd + Pb + As			6.89±0.01		2±0.02		58±0.11		
	Be + Cr + Sn + Sb + Cu + Co + Mn + Ni + V			3.48±0.11		34±0.45		80±0.33		

Notes: ND means not detected. By-gas means bypass flue gas. A_1 means that the bypass system was not used. A_2 means that the bypass system was used. Ba means the baseline test. Co means the co-processing test.

limit, indicating that the By-gas should not be discharged directly to the atmosphere.

3.3 CKD PCDD/Fs concentrations

Xiao et al. (2018) found that PCDD/Fs released in cement kiln output streams primarily migrate into flue gas, CKD, and CK. Many studies have demonstrated that co-processing does not affect CK PCDD/Fs content (Yang et al., 2012; Yan et al., 2014; Yan et al., 2018; Xiao et al., 2020). PCDD/Fs emissions in the flue gas were discussed in section 3.2. The PCDD/Fs content of the CKD samples were shown in Fig. 2(b). The PCDD/Fs concentrations were lower in the A_2-Co and C-Co samples than in the baseline samples, which was consistent results of a study performed by Xiao et al. (2018). However, the PCDD/Fs concentration were higher in the B-Co samples than that in the B-Ba samples. This was likely because the kiln had been used to dispose of MSW for a long time, which caused the PCDD/Fs concentrations to fluctuate (Yan et al., 2014). Interestingly, the PCDD/Fs content of the By-CKD was much lower than the PCDD/Fs content of the CKD. This resulted from the bypass pipe extracting little gas from the high-temperature kiln ($< 5\%$ of the total volume) (Karstensen et al., 2010), meaning that smaller amounts of PCDD/Fs would have absorbed to the By-CKD. The PCDD/Fs content of the A_2-Co CKD was 10 times higher than the PCDD/Fs content of the A_1-Co CKD, indicating that the PCDD/Fs concentration in the flue gas was decreased by the bypass system (Sutou et al., 1999).

3.4 Chlorine mass balance

Chlorine migration and enrichment are closely related to PCDD/Fs formation (Addink et al., 1996; Wikström et al., 2003; Jin et al., 2018). Chlorine was supplied to the kilns in the RM, coal, and Co-M and emitted from the CK, CKD, By-CKD, flue gas, and By-gas (Fig. 3).

In Fig. 4, RM was the primary source of chlorine in all three plants. The chlorine inputs in the RM were 77.3–259.87 kg/h. The second most important source of chlorine was Co-M, which contributed 4.86%–27.25% of the total chlorine input rate during MSW co-processing. The total chlorine output rates in the A_1-Ba, A_1-Co, A_1-Co, B-Ba, B-Co, C-Ba, and C-Co tests were 77.63, 154.47, 130.81, 183.27, 273.41, 229.55, and 249.83 kg/h, respectively, which were consistent with the input rates. CK and CKD were the primary chlorine output streams, accounting for greater than 90% of the total chlorine output rate. The chlorine input rate was clearly higher in the B-Co test than that in the other tests. RM was the primary chlorine contributor, as the chlorine input rate in the RM was 259.87 kg/h, caused the PCDD/Fs concentration to be higher in the total flue gas (flue gas plus By-gas) in plant B than in the other plants (Fig. 2(a)). The high PCDD/Fs content of the plant C CKD (Fig. 2(b)) was also related to the chlorine mass flow rate in the plant. A comparison of the contributions of the A_1-Co and A_2-Co chlorine mass flow rates (Fig. 4) indicated that the bypass system also contributed to the chlorine emissions (the bypass system in A_2-Co accounted for 7.48% of the chlorine output flow rate), indicating that the bypass system mitigated the negative effects of chlorine in the kiln.

3.5 Heavy metal distribution characteristics

The HMs input flow stream included the RM, coal, and Co-M (MSW or RDF plus IC), and the primary output flow stream included CK, CKD, By-CKD, flue gas, and By-gas, which was similar to the supply and emissions of chlorine. The detailed HMs mass flows of the three MSW co-processing techniques in the input and output streams are

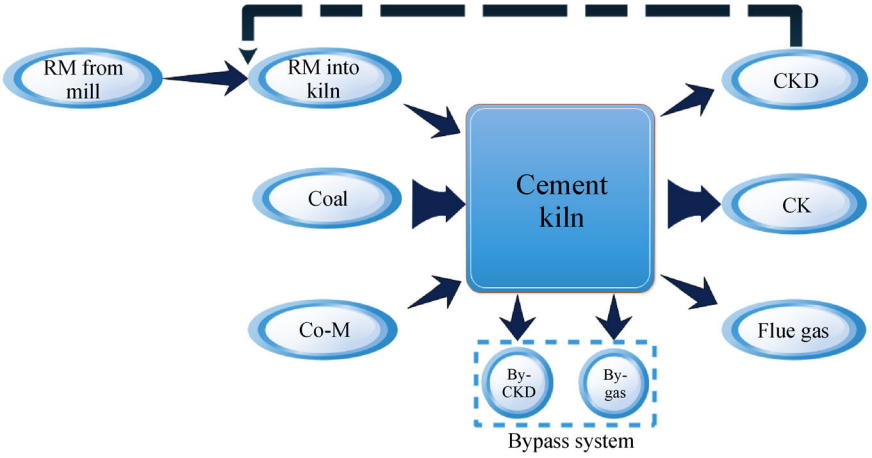

Fig. 3 Flow chart showing chlorine inputs and outputs for the cement kiln co-processing of municipal solid waste. Co-M indicates the refuse derived fuel (RDF) in plant A; RDF and inert components in plant B; and municipal solid waste in plant C. By-gas means the bypass flue gas. By-CKD means the bypass cement kiln dust.

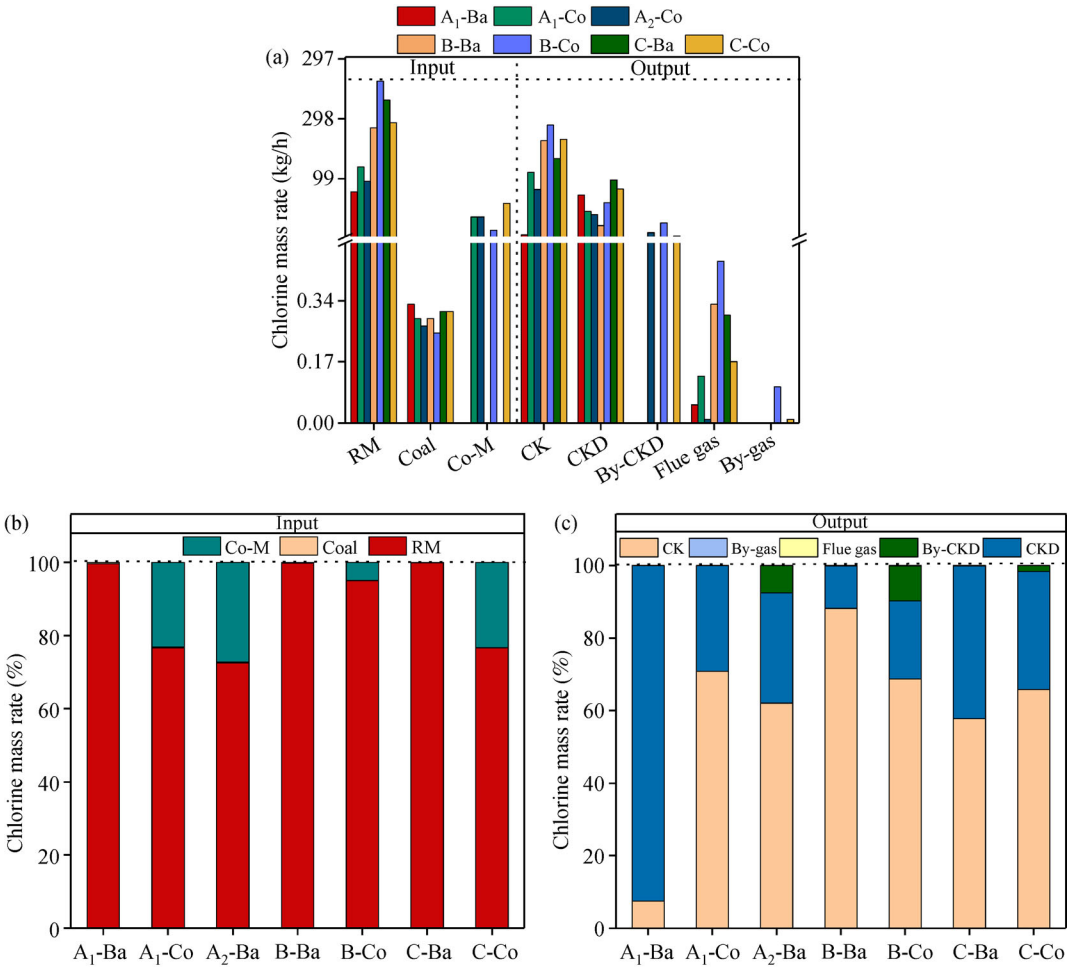

Fig. 4 (a) Chlorine mass inputs and outputs. Chlorine (b) inputs and (c) outputs for the different sources in the different plants. Co-M means refuse-derived fuel (RDF) in plant A; RDF and inert components in plant B; and municipal solid waste in plant C. A_1 means that the bypass system was not used; A_2 means that the bypass system was used; Ba indicates the baseline test. Co indicates the co-processing test. By-gas means the bypass flue gas. By-CKD means the bypass cement kiln dust.

presented in the supporting material.

RM was the primary input that contributed Hg in all three plants, and that the amounts of Hg emitted in the output streams decreased in the order of: flue gas>CKD>CK (Table S5). Differences between the Hg migration patterns in the output streams (total flue gas, total CKD (CKD plus By-CKD), and CK) for the three plants were smaller during the co-processing test than during the baseline test. This was likely because the Hg contents of the Co-M in the input streams were lower during each co-processing test than the corresponding baseline test. The RM was the primary semi-volatile HMs input stream for all three plants. The total CKD and CK were the primary contributors of semi-volatile HMs in the output streams, as only small amounts of semi-volatile HMs were emitted in the total flue gas. Be, Co, Cr, Mn, Ni, and V are low-volatility HMs. RM was the primary contributor of these HMs to the input streams in all three plants, and CK and total CKD were the primary contributors of these HMs to the output streams in all three plants, and similar results were found by Yan et al. (2015). We concluded that HMs would not cause more problems when co-processing MSW than under standard cement kiln operating conditions.

The results indicated that the RM was the primary contributor of HMs, and Co-M was not the primary contributor of HMs, in the input streams. In addition, the HMs, except for Hg, in the output stream were found primarily in the solid samples (total CKD and CK). The HM migration mechanism was not summarized above. There is a lack of a Chinese standard for HMs in hot raw meal (HRM), CKD, and By-CKD, and the behaviors of HMs in HRM, CKD, and By-CKD have not been well-characterized. Therefore, the circulating ratio concept was developed. We calculated the HMs outer-circulating ratio (OCR), inner-circulating-ratio (ICR), and bypass-circulating- ratio (By-CR) for the three plants using the equations shown below.

$$\mathrm{OCR} = \mathrm{CKD}/\mathrm{RM}, \quad (1)$$

$$\mathrm{ICR} = \mathrm{HRM}/\mathrm{RM}, \quad (2)$$

$$\mathrm{By-CR} = \mathrm{By-CKD}/\mathrm{RM}. \quad (3)$$

The range of the Hg OCR value of the three plants was 2.00–3.96, (Table 4), which is likely to be related to the physical properties of volatile Hg. The boiling point of Hg is 356.7°C (1 Pa). Cement kilns generally operate at temperatures greater than 1000°C. Hg in the RM released in the preheater and rotary kilns and then directly entered the flue gas purification system. The decreasing purification system temperature would then cause the Hg to condense and absorb to particles. Then, nearly all of the Hg would have migrated to the flue gas and CKD. Cd, Pb, and Se had higher ICR value (1.76–22.58) and By-CR value (0.40–94.17) than the OCR value (0.36–10.78), confirming that they were semi-volatile HMs and accumulated in the HRM and By-CKD (the distribution in the three plants was greater than 70%). Zhang et al. (2009) suggested that cement production and MSW co-processing can effectively immobilize semi-volatile HMs, such as Cd and Pb. This would mean that large proportions of semi-volatile HMs in co-processed MSW could not be emitted to the atmosphere. All three circulating ratios were low for the low-volatility HMs (Be, Co, Cr, Cu, Mn, Ni, Sb, and V), indicating that these HMs did not easily migrate in MSW co-processing and primarily accumulated in the CK. We also assessed Mo volatilization in cement kilns. The ranges of Mo By-CR and ICR values in the three plants were 3.88–5.87 and 1.28–2.05, respectively, indicating that Mo was a semi-volatile HM in terms of the cement production process. Some "low volatility" HMs had high circulating ratios (Table 4), which showed volatile compounds such as $CuCl_2$ and $SbCl_3$ were present in the input stream. The results concluded that the co-processing MSW did not obviously increase the HMs circulating ratios.

The HMs distributions in the cement kiln materials were closely related to the chemical forms of the HMs compounds, but this was outside the scope of the present study.

3.6 Leaching of heavy metals from cement products

HMs concentrations in the leachates of cement products were lower than that in the technical specification for co-processing of solid waste in cement kiln, released by Standardization Administration of China in 2014 (Table S6). The co-processing MSW had little effect on the compositions and physical properties of the RM and CK, which met the requirements of Portland Cement Clinker, released by Standardization Administration of China in 2008 (Table S7). These findings indicated that the CK produced by the co-processing MSW can be used in the subsequent production of cement.

4 Conclusions

Tests were performed by using three common cement plants in China to assess the effects of co-processing MSW in cement kilns.

The MSW pre-treatment results indicated that the three MSW disposal methods could produce Co-M. The RM replacement rates for the plant B and C co-processing were

Table 4 Heavy metal outer-circulating ratio (OCR), inner-circulating ratio (ICR), and bypass-circulating ratio (By-CR)

HM	A_1-Ba-OCR	A_1-Co-OCR	A_2-Co-OCR	A_2-Co-ICR	A_2-By-CR	B-Ba-OCR	B-Ba-ICR	B-Co-OCR	B-Co-ICR	B-By-CR	C-Ba-OCR	C-Ba-ICR	C-Co-OCR	C-Co-ICR	C-By-CR
Be	1.59	0.82	2.86	1.29	2.29	1.01	0.83	0.90	0.90	0.79	1.20	0.67	0.87	0.80	0.63
V	0.86	0.70	2.17	1.27	1.94	1.03	1.20	0.98	1.24	1.21	1.21	1.21	1.26	1.15	1.16
Cr	0.81	0.78	1.42	1.19	1.78	0.85	0.92	0.54	0.79	0.90	1.00	1.30	1.06	1.03	1.30
Mn	0.17	0.71	1.64	1.28	1.75	0.78	1.22	0.82	1.30	0.77	0.82	0.93	1.13	1.05	2.57
Co	1.12	1.02	2.46	1.26	2.07	0.94	1.16	0.93	1.24	0.84	1.19	1.31	1.15	1.28	1.24
Ni	1.56	1.01	1.72	0.91	1.48	0.69	0.74	1.68	1.46	1.15	1.12	1.38	1.19	1.22	1.36
Cu	1.22	1.08	2.85	1.94	8.91	1.06	1.38	1.28	1.61	1.65	1.18	1.40	1.06	1.31	1.73
Zn	1.42	1.90	2.38	1.27	2.74	1.96	0.52	3.52	1.78	1.52	1.18	1.20	1.07	0.99	1.28
Se	1.16	0.92	2.16	6.53	8.89	0.90	6.39	1.13	6.28	24.30	1.19	19.19	1.67	22.58	94.17
Mo	0.90	0.80	2.17	1.43	5.87	0.82	1.28	1.01	1.65	5.77	1.12	1.94	1.24	2.05	3.88
Cd	0.87	1.04	6.58	6.53	93.21	0.85	2.75	0.36	5.95	8.36	1.54	1.76	1.17	1.83	4.24
Sb	2.91	1.04	7.14	1.91	15.36	1.49	0.91	2.07	1.84	1.48	1.25	1.16	1.04	0.74	1.00
Hg	2.00	3.19	3.30	0.00	4.00	3.57	1.45	4.04	0.65	0.28	2.26	0.85	3.96	0.34	1.07
Tl	6.30	5.51	0.07	0.06	0.00	2.70	1.19	3.26	0.21	0.29	14.48	0.31	13.30	0.20	1.33
Pb	1.67	1.20	10.78	6.09	0.40	0.94	2.85	1.11	7.58	16.10	1.65	2.34	1.43	3.12	7.62

Notes: A_1 means that the bypass system was not used. A_2 means that the bypass system was used. Ba indicates the baseline test. Co indicates the co-processing test.

1.15% and 1.41%, respectively. The RDF produced in plant A accounted for 54% of the MSW, and the lowest calorific value was 1472.89 kcal/kg. The RDF produced in plant B accounted for 39.2% of the MSW, and the lowest calorific value was 1156.58 kcal/kg. The effective CG components produced in plant C accounted for greater than 80% of the MSW.

During the CK production during test A_2-Co, the SO_2 concentration was greater than the relevant limit. This suggested that the RDF feed rate needed to be decreased. The TOC concentration in the B-Co flue gas was greater than the relevant standard, suggesting that the RDF size should be decreased, and that the kitchen waste should not be fed into the raw mill. The TOC concentration in the C-Co flue gas was greater than the relevant standard, suggesting that the CG feed rate should be decreased. Besides, the co-processing MSW did not interfere with HMs migration. The circulating ratios indicated that Mo acted as a semi-volatile HM in the cement kilns. The PCDD/Fs emissions in the By-gas for all three co-processing methods were greater than the relevant limits. The TOC concentrations in the B-Co and C-Co By-gas samples were greater than the relevant limit. This suggested that the By-gas could be introduced to the primary flue system for disposal. The chlorine mass balance indicated that CK and CKD were the primary output streams containing chlorine, which may result from the large amount of chlorine in the RM.

Co-processing MSW in cement kilns in China has not allowed "zero pollution" MSW disposal to be achieved. Problems caused by co-processing MSW in cement kilns need to be further addressed in future studies.

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Author contributions All authors contributed to writing this manuscript. All authors have approved the final version of the manuscript.

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