

Use of non-recycled plastics and paper as alternative fuel in cement production

A.C. (Thanos) Bourtsalas^{a, *}, Jiao Zhang^a, M.J. Castaldi^b, N.J. Themelis^a, Alexandros N. Karaiskakis^c

^a Earth Engineering Center, Columbia University, New York, NY, 10027, USA

^b Earth Engineering Center, City College of New York, New York, NY, USA

^c Department of Chemical Engineering, The City College of New York, CUNY, New York, NY, 10031 USA

ARTICLE INFO

Article history:

Available online 3 February 2018

Keywords:

Cement production
Energy
Non-recycled plastics
Plastic waste
Paper waste
Alternative fuel
Environmental effects

ABSTRACT

The global cement industry produces over four billion tonnes annually. In recent years, the quest for increasing energy efficiency has led this industry to test and use “waste” materials as alternative fuels (AF). The objective of this study was to examine the use, as alternative fuel, of the shredded non-recycled plastics and paper residue (NRPP) of a materials recovery facility at the Balcones plant in San Antonio, Texas; this material has a Lower Heating Value (LHV) of about 17 MJ/kg and is called “engineered fuel” (EF). The results showed that EF consists of mostly paper fiber, a biogenic material, so its use helps to decrease CO₂ emissions from cement production. In the US, on an average about 4.3 MJ of thermal energy are used to dry, decompose, and sinter the carbonate minerals to produce one kg of “clinker” that is then ground to cement powder and mixed with other compounds. If the maximum amount of EF that can be used by the U.S. cement industry (83 million tonnes of cement per year) were to be separated at Material Recovery Facilities and used as alternative fuel in cement kilns, the diversion of non-recycled paper and plastics from landfills would amount to 17.7 million tonnes of EF. The general methodology used for the Life Cycle Assessment (LCA) is *Eco-indicators 99* that uses the SimaPro database and four scenarios were assessed. The corresponding chemical structure of the EF was calculated and the associated emissions during combustion were determined. The laboratory analyses determined the average makeup of the sampled fuels consisted of 20% fossil based carbon and 80% biogenic. The use of EF in the cement industry reduces greenhouse gas emission by up to 3 tonnes of CO₂ per tonne of EF used in place of a high-quality coal. The study also found that use of EF in cement production has no adverse effect on the stack emissions of cement plants, nor on the quality of cement produced. The mercury concentration in the stack gas of U.S. cement kilns is well below the U.S. standard and the total dioxin emissions of all cement plants amount to only 0.05% of the dioxins emitted by all U.S. sources. Furthermore, the cement process incorporates the residual ash in the EF into the final clinker, thus resulting in both energy and materials recovery.

© 2018 Elsevier Ltd. All rights reserved.

1. Introduction: characteristics of global and US cement production

The cement industry is one of the largest in the world and, typically, about 3–5 MJ of thermal energy are used to dry, decompose, and sinter the carbonate minerals to produce one kg of “clinker” that is then ground to cement powder, and mixed with other components. For the first time in history, the annual output of cement exceeded 4 gigatonnes (Gt) in 2015. China, with an output

of 2.4 Gt or 57.3% of the total global production is the world's leading producer, followed by India 6.6% of world production and the U.S. (2.0%), as presented in Fig. 1.

In the period between 1993 and 2005, U.S. cement production increased from 67 million tonnes to a maximum of 99 million tonnes in 2005. It decreased to a low of 65 million tonnes in 2009, due to the economic crisis and then recovered to 83 million tonnes by 2015. The estimated clinker capacity for 2015 was 106 million tonnes of clinker. Therefore, the average energy consumption was calculated at about 4.5 MJ/kg of cement in 2013, and was about the same since 1993 as presented in Fig. 3 (USGS, 1993–2016; Bye, 2011; Marceau, 2006).

Fossil fuels have been used in cement production but the

* Corresponding author.

E-mail address: ab3129@columbia.edu (A.C. (Thanos) Bourtsalas).

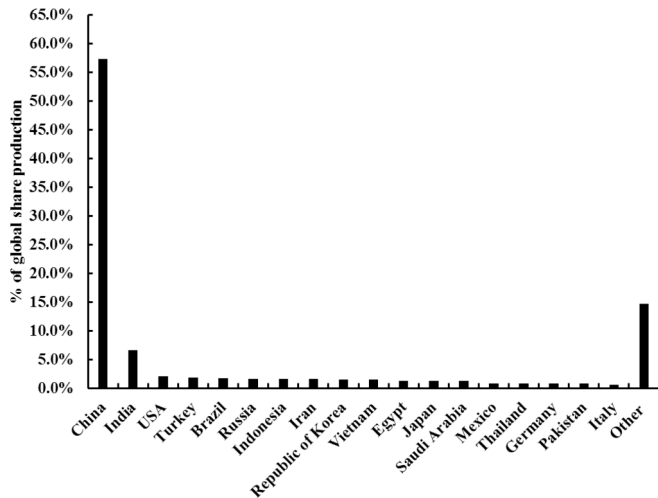


Fig. 1. World cement production in 2015, total 4.1 billion tonnes (USGS, 1993–2016).

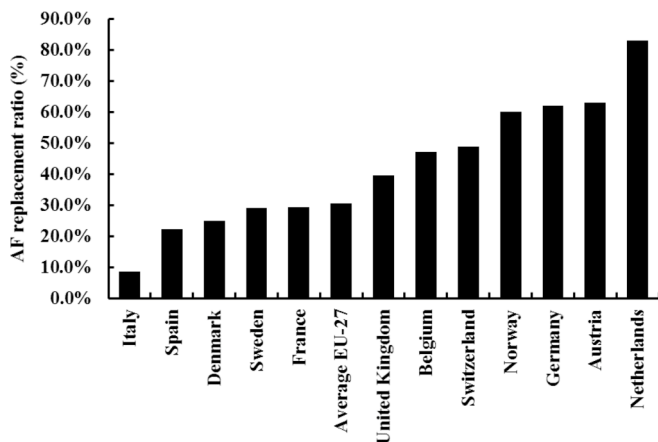


Fig. 2. Replacement ratios of fossil fuels with alternative fuels (AF) in European countries. The average for EU-27 is 30.5% (Zhang, 2013).

increasing pressure for lower operating cost and reduced environmental impact has led the global cement industry to use “waste” materials as alternative fuels (AF). AF used in cement production include a variety of waste fuels, such as automobile shredder residue (ASR), carpet residue, textiles, wax residue, meat and bone meal, landfill gas, and fuels derived from municipal solids waste (MSW) streams. It is desirable for the properties of AF to be close to petcoke since the process has been optimized for that fuel. Petcoke is a solid carbon material, which is a by-product of the oil refining process. Table 1 (Nasrullah et al., 2015; Gallardo et al., 2014; Rada and Ragazzi, 2014; Zhang, 2013; Zhou et al., 2013; Patel et al., 2012; Ahn et al., 2013; Garg et al., 2007; Skodras et al., 2006; Grammelis et al., 2009; Piao et al., 2000; Dai et al., 2001; Conesa et al., 2004; Wan et al., 2008; Hassan et al., 2009; Scala and Chirone, 2004; Senneca et al., 2002; Zou et al., 2007; Ayllon et al., 2006; Kaantee et al., 2004; Wang et al., 2004; Trent et al., 1982; Brady and Hatch, 1997; NETL, 2012; Littlefield, 2009; Aho and Ferrer, 2005; Borgianni et al., 2002; Liu et al., 2001; Jannelli and Minutillo, 2007) shows that many of the AF materials meet one or two specifications, e.g., moisture and ash content, but not others, such as Cl concentration and heating value. Tires and Tire Derived Fuels (TDF) have a similar high heating value to petcoke (34.4–36.2 MJ/kg) while refuse derived fuels (RDF) are lower

in heating value than petcoke and coal.

The use of waste materials in cement plants has increased in recent years. In 2012, over two thirds of the nearly 270 cement plants in the European Union used some alternative fuels. The average use of AF was about 30% of the total thermal energy, with the highest levels of 62% in Germany, 63% in Austria, and 83% in the Netherlands (Fig. 2 (Zhang, 2013)).

The typical heating values of the various fuels used in the US cement production are shown in Table 2 (US EPA, 2008; IPCC, 2006; United Nations, 2010). The units of all fuels are expressed in MJ/kg. The total energy consumption of the US cement industry is calculated at 3.8 MJ/kg of cement and 4.2 MJ/kg of clinker for the year 2014 and is slightly reduced as compared to 1993, which was about 4.3 MJ/kg of cement and 4.4 MJ/kg of clinker. Fig. 4 shows the kinds of fossil and alternative fuels used by the U.S. cement industry, in the period 1993–2014, expressed as a percent share of fuels in the total energy consumption. In the U.S., the use of AF (tires, RDF, wood, liquid wastes, etc.) in the cement industry increased from 8.7% to 18.9% of the total energy consumption (USGS, 1993–2016). The AF used are divided into three big categories: tires, other solid waste and liquid waste. Both “other solid waste” and “liquid waste” may include several materials and their LHV varies widely. The use of “other solid wastes” in the U.S. cement industry increased from 90 thousand tonnes in 1993 to 1.3 million tonnes in 2014 which, at an assumed LHV of 18 MJ/kg, represented about 39.4% of the AF energy used in cement production (7.4% of the total fuel used) (USGS, 1993–2016). The liquid waste, with an assumed LHV of 15 MJ/kg, contributed to about 39.6% of the AF energy used in cement manufacturing (7.5% of the total) and tires, with a LHV of 32.6 MJ/kg, contributed at about 21.1% to the AF energy produced for cement production (3.9% of the total). The use of coal, with LHV of 26.3 MJ/kg, dramatically decreased from 82.8% in 1993 to 56.7% in 2014 of the total energy used in the US cement industry. The contribution of petcoke (34 MJ/kg) to the total energy used was slightly increased from 14.1% in 1995 to 17.1% in 2014. However, there is a tendency of the cement industry to move away from the use of fossil fuels and use AF instead. This is associated with the observed steady increase of the ‘other solid waste’ fuels use since 1993 (USGS, 1993–2016).

However, there is often a negative public perception for the use of AF in combustion processes, mainly associated with the chlorine content of these fuels and the formation of dioxins and furans during combustion. Dioxins and furans and other organic pollutants of environmental concern emissions are negligible, mainly associated with the long residence time of the fuel to the cement kiln and the level of temperature inside the kiln. An inventory by Columbia University (Dwyer and Themelis, 2015) of all controlled (i.e. industrial) and non-controlled (i.e. landfill fires, back yard burning, etc.) sources of dioxins in the U.S. showed that the total annual dioxin emissions of the U.S. cement industry amounted to 18 g TEQ, corresponding to 0.05% of all U.S. sources of toxic dioxins (Dwyer and Themelis, 2015). Also, a survey (WBCSD, 2006) of nearly 2200 worldwide dioxin emissions data, from 1990s to 2005, of cement kilns co-combusting AF concluded that the use of AF had a negligible effect on dioxin emissions; in some cases where AF contained trace amounts of heavy metals, they were stabilized in the clinker or entrapped by the Air Pollution Control (APC) systems, thus resulting in negligible difference of heavy metal emissions between plants combusting only fossil fuels and those co-combusting AF (Kara, 2012; Schneider et al., 2011; Shih et al., 2005; WBCSD, 2006).

The energetic and environmental impacts of using AF for cement and clinker production have been extensively examined with Life Cycle Analysis (LCA) models, laboratory studies and industrial surveys (Kajaste and Hurme, 2016; Gursel et al., 2014; Li

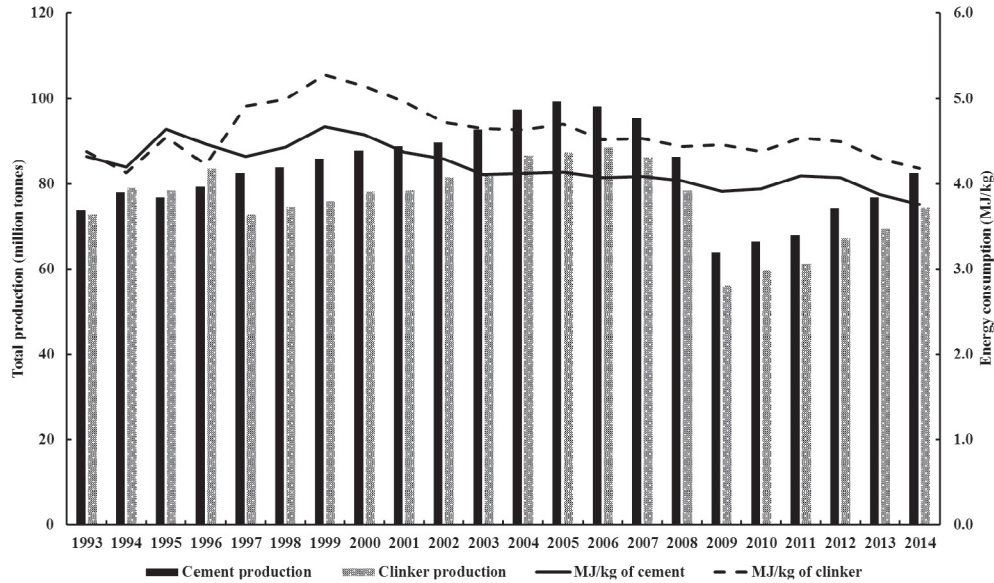


Fig. 3. Clinker and cement production (primary axis: million tonnes) and energy consumption (secondary axis: MJ/kg), U.S. 1993–2014 (USGS, 1993–2016).

Table 1
Proximate and ultimate analysis and heating values of fuels used in cement production. Notes to Table 2 shown in following page: a) Pine wood and waste wood, b) Refuse Derived Fuel from Municipal Solid Waste, c) Tire Derived Fuel, d) Meat and Bone Meal, e) Poly-Ethylene, f) Poly-Vinyl-Chloride, g) American coal, (*) calculated. The Low Heating Values (LHV) are frequently used in Europe.

Fuel Type	Proximate Analysis (wt. %, as received)				Ultimate Analysis (wt. %, dry basis)						Alkali		LHV	HHV		
	Moisture	Volatile Matter	Fixed Carbon	Ash	C	H	N	S	Cl	O	Na	K	MJ/kg	MMBTU/ton	MJ/kg	MMBTU/ton
Wood^a (12), (18)	5.6–6.3	69.5–78.5	15–16.1	0.5–8.8	46–51.3	5.7	0.07	0.01	≈0	35.4–36	≈0	≈0	17.1*–22.5*		18.6–23.9	
RDF^b (4)–(11), (14), (19), (20), (21), (31), (32)	3.7–20	71.9–76	3.9–13.2	10.2 –13.8	41.7 –50.2	4.4 –7.8	0.75 –1.65	0.1 –0.76	0.7 –1.13	28.5 –36.3	0 –0.93	0 –0.37	15.9*–17*	13.7–14.6	17.5–18.4	15.1–15.9
Tires (15), (16), (23), (28)	0.7–4	63.8–68.7	24–31.1	2.2–8.2	76.7 –89.4	7–7.8	0.2–0.5	0.8–2.2	≈0–0.1	0.4–4.5	0–0.9	0–1.1	32–35.6 28.4–30.7		34.6*–37.3 29.8–32.1	
TDF^c (19), (20)	0.9–1.9	63.4–64	30.4–30.7	3.3–4.4	83.8 –86.7	6.9	0.3–0.6	1.9–2	≈0	0.9–2.3	n/a	n/a	31.8–36.8 27.4–311.7		33.5*–38.4* 28.9–33.1	
Pecan Shells (28)	14.6	n/a	n/a	2.32	46.84	5.41	0.44	n/a	n/a	n/a	n/a	n/a	18.2*		19.8	
MBM^d (22), (23)	1.4–8.1	64.5–79.7	7.2–9.7	10.4 –28.3	42.1 –55.7	5.8–8	7.2–8.9	0.05 –0.4	0.2	15.3 –38.4	1.5	0.3	19.6–28.8*	16.9–24.8	21*–30.6	18.1–26.4
PE^e (20), (24)	≈0 –0.17	99.8–100	≈0	≈0 –0.06	86	14	≈0	≈0	≈0	≈0	n/a	n/a	44.6 38.4		44.9* 38.7	
PVC^f (13), (24), (30)	≈0–0.2	85.9–91	6.3–9	1–7.6	35.9–38	4.4–5	0–0.11	≈0	57	≈0	n/a	n/a	17.1*		18.1	
Sewage Sludge (19), (23)	5.2–5.6	40.5–85	5–10.1	17.9 –29.5	36.4 –40.5	4.7–7	0.84 –5.0	0.1–0.6	≈0–1	22	≈0	≈0	9.3–15.8 8.0–13.6		10.4*–17.5 9.0–15.1	
Pet. Coke (21), (23)	0.8–1.5	7.6–10	88.6–89.6	0.5–1	89.5 –92.7	2.4 –3.7	1.2–1.7	1.5–4	≈0	1.1–1.2	≈0	≈0	33.5–35.4* 28.9–30.5		34.4*–36.2 29.6–31.2	
Coal^g (19, 25–27)	1.1–3.3	23–35.3	44.2–66.8	6.4 –15.5	65.3 –80.9	3.7 –5.1	1.2 –1.41	0.6–5.5	≈0 –0.33	5.9 –12.5	0 –0.03	–0.19	25.4–31.8* 21.9–27.4		26.3*–32.9 22.7–28.3	

et al., 2014; Valderrama et al., 2012; Chen et al., 2010; Huntzinger and Eatmon, 2009; Galvez-Martos and Schoenberger, 2014; De Queiroz Lamas et al., 2013; Strazza et al., 2011; Rovira et al., 2010; Genon and Brizio, 2008; Mokrzycki et al., 2003; VDZe.V. V.D.Z., 1996; Karstensen, 2008; Dwyer and Themelis, 2015; Kara, 2012; Schneider et al., 2011; Shih et al., 2005; WBCSD, 2006; European Commission, 2003; Montejo et al., 2011). However, there are no

studies on the use of non-recycled plastics and paper (NRPP) as AF in cement production and the associated potential of advancing waste-to-energy in a nation. The objective of this study is to fill this gap. Data were obtained from the Balcones cement plant in San Antonio, Texas, which receives engineered fuel (EF) comprised of NRPP from a Materials Recovery Facility located nearby.

Table 2
LHV of fuels used in U.S. cement industry [33–35].

Fuels	LHV (MJ/kg)
Coal	26.3
Petcoke	34.0
Tires	32.6
Coke from coal	28.2 (IPCC, 2006)
Fuel Oils	40.0 (IPCC, 2006)
Natural Gas	53.5 (United Nations, 2010)
Other solid waste	18.0
Liquid waste	15.0

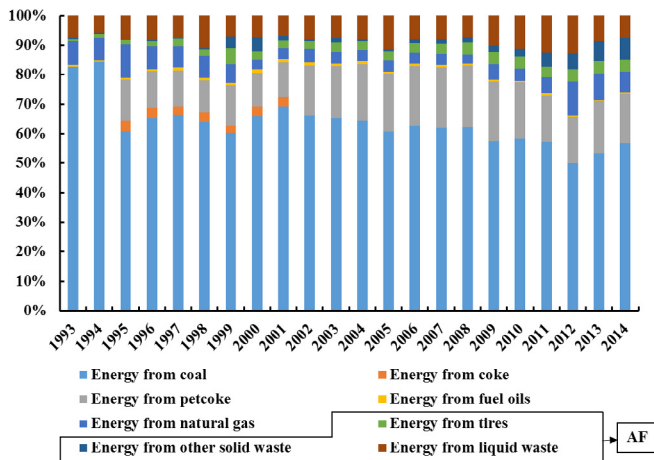


Fig. 4. Energy use by different fuels in U.S. cement industry, 1993–2014 (USGS, 1993–2016).

2. Characteristics of Balcones plant and alternative fuels used

The Balcones plant operates with a rotary kiln, 54 m long, and a kiln, where secondary combustion takes place, 74 m long. Fig. 5 depicts a schematic of the two kiln lines, both having the flow path for solids and gases. The rotary kiln is equipped with a 5-stage preheater precalciner with raw gas temperatures of about

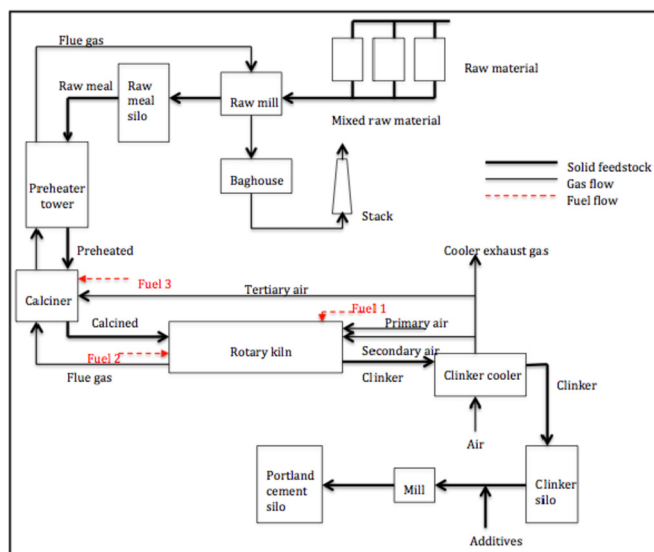


Fig. 5. Flow diagram of cement process. Fig. 5 depicts a schematic of the two kiln lines, both having the flow path for solids and gases (Zhang, 2013).

310–320 °C. The calciner is designed for a long retention time to ensure that AF are effectively combusted and that CO and VOC emissions are kept to the minimum. The air pollution control (APC) system of Balcones includes selective non-catalytic reduction for NO_x emissions (SNCR; ammonia injection), and baghouse fabric filter for particulate matter (PM) control.

At the Balcones facility, the typical AF used are: “Engineered fuel” (NRPP), wood, tires and pecan shells. EF is provided to the cement plant by a materials recovery facility (MRF) in San Antonio where source-separated recyclables are sorted out to marketable paper, plastic, metal, and glass. The Balcones MRF was built in 2012 on a 10.6-acre site with a capital investment of \$ 24 million or \$278 per ton of MSW. The gate fee of the facility is about \$35 per ton of MSW. The total inbound tons of all residential, commercial, direct feed fiber and plastics averages 7200 tons per month of which about 4000 tonnes are residential waste. The residential single stream is being processed at 25 tonnes per hour and the commercial single stream is being processed at 15–17 tons per hour. Balcones is currently operating a single 8.5-h shift Monday through Saturday. In 2013, the production of EF amounted to about 3000 tonnes (Zhang, 2013).

The Balcones process involves mixing of the as-received MSW to specific plastic-to-fiber ratios depending on waste availability, process requirements, and customer needs. The mixed MSW is shredded before being fed at a controlled rate into two parallel fuel extruders with adjustable dies that control the extrusion dimensions. The fuel extrusion process is primarily accomplished using pressure. Heat can also be added with the use of resistance heating elements. Once extruded, the fuel pellets fall onto a conveyor and stored in truck containers ready for delivery.

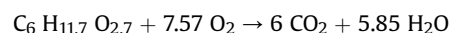
The residue consists primarily of non-recycled plastics and paper fiber (NRPP) with a calorific value of 17.5 MJ/kg, which is similar to the US average of ‘other solid fuels’ (18 MJ/kg). It is shredded and transported by truck to the cement plant. A typical flow diagram of the NRPP production process is shown in Fig. 6.

The monthly composition of the AF combusted at the Balcones plant, in 2013, is presented in Fig. 7. In February 2013, the facility reached a monthly record of NRPP (12.5% of total fuel used) used as AF. Also, in April 2013, Balcones site reached a peak in the use of energy from AF (38.9% of total fuel used), which was significant higher than the US average usage of AF (19.7%).

3. LCA study and calculation of greenhouse gas effects of using NRPP

The emissions of CO₂ by the U.S. cement industry in 2013 were calculated to be about 60.7 million tonnes, i.e., 0.87 tonnes of CO₂ per tonne of clinker produced (USGS, 1993–2016). Table 3 compares the U.S. MSW and cellulose (paper fiber) compositions. The molecular weight and the heat of formation of the various compounds were obtained from HSC 9.1 software. The corresponding chemical structure of the first three materials was calculated using the atomic composition of each material (Zhang, 2013). The most biogenic material, poplar wood, contains 4.2 atoms of oxygen for every 6 atoms of carbon; while the highest fossil-based material, TDF, contains very little oxygen.

The combustion of the NRPP fraction is associated with the following reaction:



which is a highly exothermic reaction with 23.8 MJ of energy released/kg of fuel, which is higher than the calculated value of MSW, 17.9 MJ/kg, as attenuated. However, the reported heating value of the NRPP used at the Balcones plant was 17.5 MJ/kg. The

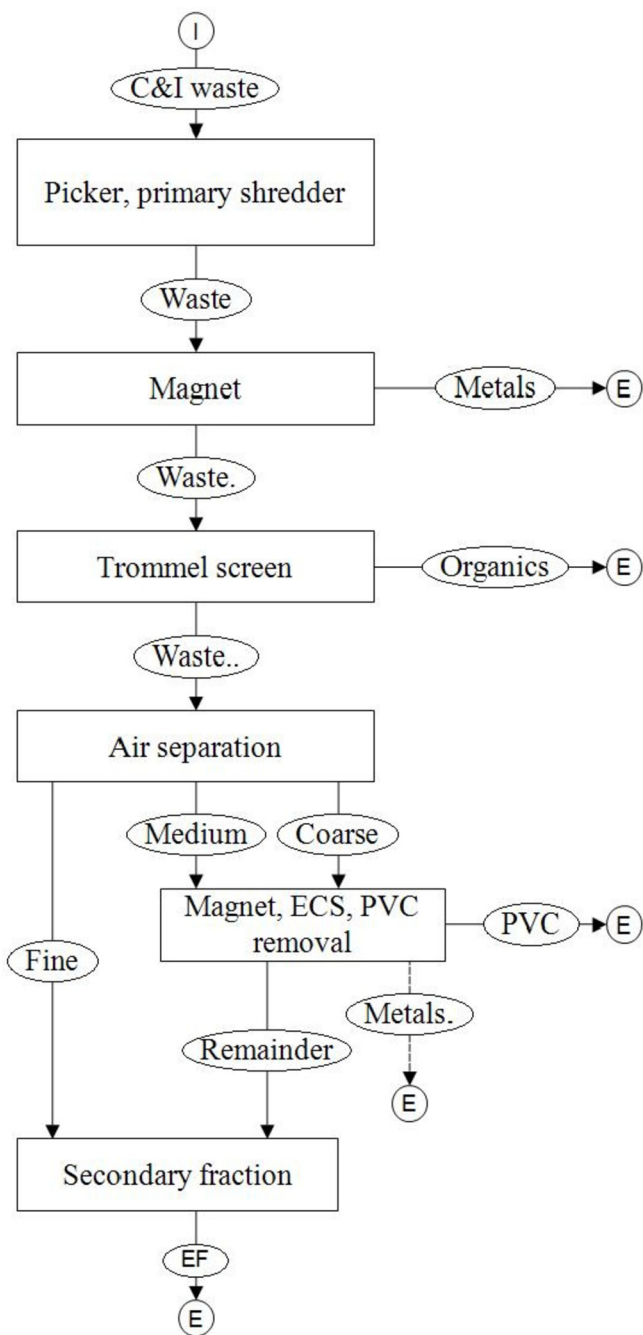


Fig. 6. Production process of EF, which consists of non-recycled plastics and paper fiber (NRPP), used at Balcones plant. The input waste to the Materials Recovery Facility (MRF) for the recovery of EF-NRPP is Commercial and Industrial Waste (C&I). In 2013, the total inbound tons of all residential, commercial, direct feed fiber and plastics averages 7200 tons per month of which about 4000 tonnes are residential waste. In the same year, the EF production was about 3000 tons.

losses are mainly associated with the moisture and the impurities contained in the NRPP fraction.

The CO₂ emissions were calculated by the above chemical equation, which gives 1.93 kg of CO₂ per kg of NRPP. MSW combustion is associated with 1.68 kg of CO₂ and tires with 3.10 kg of CO₂ per kg of fuel, as presented in Table 3. Also, the combustion of pet coke is associated with 3.56 kg of CO₂ per kg of pet coke combusted, as a result of the significant higher amount of carbon contained in petcoke compared to NRPP/RDF, MSW and other waste

fuels, except for waste tires.

The general methodology used for the Life Cycle Assessment (LCA) is *Eco-indicators 99* that uses the SimaPro database. Four different scenarios were considered to determine the carbon dioxide emissions of using NRPP in place of coal in the cement production process:

- **Scenario 1 (best case):** 75% of the thermal energy is derived from NRPP and 25% from coal.
- **Scenario 2:** 50% of the energy is derived from NRPP and 50% from coal.
- **Scenario 3:** 25% of the energy is derived from NRPP and 75% from coal.
- **Scenario 4 (worst case):** 100% of the energy is derived from coal.

The functional unit was 1 tonne of clinker production and the assumptions made for the CO₂ calculation were as follows:

- Total energy demand of 4500 MJ/tonne of clinker produced.
- Heating value of 26.3 MJ/kg, for coal, and 17.5 MJ/kg, for NRPP.
- Transport distance from MRF to cement plant of 50 km, using 7.5–16-tonne truck.
- Transport distance from coal mine to cement plant: 300 km via rail.
- 80% of carbon in NRPP is biogenic (paper fiber, etc.) and 20% fossil based (non-recycled plastics).
- The carbon-based compounds in NRPP are diluted with 20% moisture plus non-combustible materials.

The alternative fuel used was sampled and analyzed at EEC| CCNY laboratory using standard ASTM protocols. The analyses determined the average makeup of the sampled fuels consisted of 20% fossil based carbon and 80% biogenic. (Zhang, 2013).

Since the ash of both NRPP and coal is absorbed in the clinker the boundaries of the LCA study are defined from the production of the fuel input to the cement production process and the CO_{2-eq} emissions were calculated. In accordance with the international protocol (IPCC, 2006), only the fossil-based CO₂ emissions are considered as contributors to the greenhouse gases (GHG) causing climate change. The GHG emissions are based on the SimaPro database and the Ecoinvent program and U.S. input-output database for coal mining, production of NRPP (electricity from waste allocation from Simapro), transportation (coal freight and lorry for coal ash and NRPP), and landfill avoidance for NRPP.

For the production of NRPP-EF, the energy consumption was assumed to be 2580–3840 MWh/year. The annual energy costs were calculated to be \$180,000 – \$192,000/year at an electricity rate of 5–7 ¢/kWh. At a production rate of 36,000 tonnes of NRPP-EF per year, the energy intensity of producing NRPP-EF is estimated at 864–1272 kWh of electricity per ton of NRPP-EF produced. In this case, the energy cost is about \$5/tonne of EF (Zhang, 2013). The estimated electricity consumed was used as an input to the Ecoinvent and U.S. input-output database to obtain the CO_{2-eq} emissions per ton of EF (Cleary, 2009).

The results of the LCA analysis are shown in Table 4. It is assumed that, when the NRPP is not used in cement production, it is disposed in a sanitary landfill where 75% of the methane (CH₄) generated by the organic fraction of the NRPP is captured and 25% is emitted to the atmosphere (Zhang, 2013). Under these circumstances, the SimaPro LCA program assigns an additional GHG benefit of 1.31 kg CO₂ per kg of NRPP used in cement production.

The use of one tonne of NRPP in the cement production results in the reduction of about 1.6 tonnes of CO₂. The beneficial effect of using NRPP in place of coal, when the avoided landfill emissions are

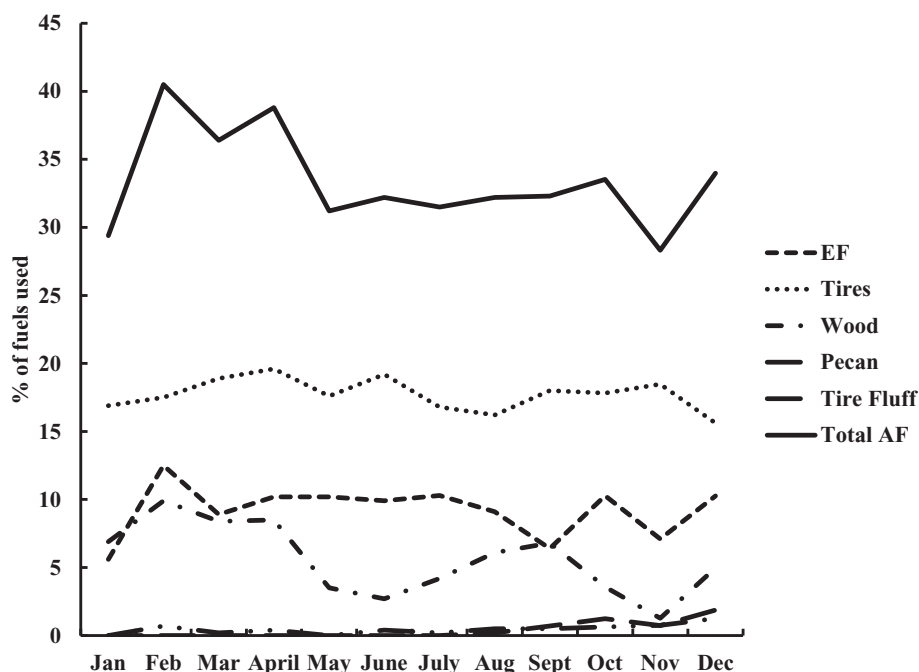


Fig. 7. Average monthly composition of AF used at the Balcones plant. Data from Jan–Dec 2013

Table 3

C, H, and O concentration in five dry materials and petcoke coal.

Composition	Tire derived fuel	RDF and NRPP	U.S. MSW	Poplar wood	Cellulose	Petcoke
%C	85.20%	48.80%	38%	47.50%	44.40%	90.00%
%H	7.30%	7.80%	5.21%	6.10%	6.20%	3.00%
%O	0.50%	29.20%	33.33%	44.50%	49.40%	1.20%
All other (%)	7.00%	14.20%	23.96%	1.90%	0.00%	5.8%
Calculated C–H–O structure	$C_6 H_{6.2} O_{0.03}$	$C_6 H_{11.7} O_{2.7}$	$C_6 H_{10} O_4$	$C_6 H_{9.2} O_{4.2}$	$C_6 H_{10} O_5$	$C_6 H_{2.4} O_{0.06}$
Formula weight (g/mol)	78.79	127.06	146.14	148.53	162.14	74.08
Carbon emissions during combustion (kg/kg of fuel)	3.10	1.93	1.68	1.66	1.51	3.56

Table 4

LCA calculation of GHG emissions for four EF-coal combustion scenarios for functional unit: 1 tonne of clinker.

	Scenario A: 75% EF		Scenario B: 50% EF		Scenario C: 25% EF		Scenario D: 0% EF
	EF	Coal	EF	Coal	EF	Coal	Coal
Energy needed MJ/tonne clinker	3375	1125	2250	2250	1125	3375	4500
Kilograms fuel per tonne clinker	193	43	129	86	64	128	171
Mining of coal, kg CO ₂ /tonne clinker		4.3		8.6		12.8	17.1
Production of EF, kg CO ₂ /tonne cl.	0.25		0.17		0.08		
Coal transport, kg CO ₂ /tonne clinker		0.5		1.1		1.6	2.1
EF transportation (kg CO ₂ /tonne clinker)	0.03		0.01		0.008		
Combustion emissions, kg CO ₂ /tonne clinker	372	153	249	306	124	456	609
Subtotal kg CO ₂ /ton clinker	372.3	157.8	249.2	315.7	124.1	470.4	628.2
Total EF+coal, for scenario, kg CO ₂ /tonne clinker	530.1		564.9		594.5		628.2
Kg CO ₂ avoidance due to EF use in cement	–98.1		–63.3		–33.7		0
Total GHG including landfill, kg/tonne clinker.	–351.1		–232.3		–117.5		0

included in the comparison of GHG emissions, results in an additional reduction of 1.3 tonnes of CO₂ per tonne of NRPP, making a total of about 2.9 tonnes of CO₂.

4. Limits for AF use for replacing fossil fuels in cement production

One of the basic specifications of the characteristics of the feedstock to the cement kiln is the maximum amount of chlorine content, which should be <0.7%. Higher amounts of chlorine may

result in the excessive formation of salts (cement kiln dust) that are volatilized in the kiln and then solidify and can foul and corrode the downstream unit (in the direction of gas flow) of the calciner (Ahling, 1979). This section discusses the corresponding upper limit of AF that can be used to replace fossil fuels in cement production.

The data for the chlorine content of different fuels were collected from different sources and small discrepancies were noted. The results are shown in Table 5. Coal and petcoke have the lowest chlorine content. The chlorine in petcoke is about 0.005–0.032%. There are high chlorine coals (>0.3%) produced in

Table 5
Chlorine contents of different fuels.

Fuel type	Chlorine (%)			
Coal	0.04 (CIWMB, 1992)	0.01–0.03 (DOE's)	0.01–0.2 (National Research Council, 2007)	0.12–0.54 (Dept. of Energy, 1993)
Petcoke	0.005–0.032 (DOE's)			
MSW	0.3–0.8 (CIWMB, 1992)	0–1.56 (0.726) ^b (Valkenburg, 2008)		
RDF		0.36–1.29 (0.7) ^b (European commission, 2003)		
Wood pellet	0.010–0.126 (0.048) ^b (Obernberger and Thek, 2004)	0.01–0.05 (0.02) ^b (Vassilev, 2010)		
Tires	0.07–0.2 (CIWMB, 1992)	0.15 (Rubber manufacturers association, 2016)		
EF	0.74 ^a			

^a Balcones Plant.

^b Number in parenthesis is the mean value.

Illinois, but the usual chlorine found in coals ranges between 0.01 and 0.04%. Wood pellets (0.01–0.126%) and tires (0.07–0.2%) have higher chlorine content than coal and petcoke, but lower than RDF (0.36–1.29%). MSW is reported to range from 0.3 to 1.56% but the typical range of the U.S. MSW is 0.4–0.6% and of the Engineered Fuel- NRPP (0.74%) (CIWMB, 1992; Rubber manufacturers association, 2016; DOE's; National Research Council, 2007; Dept. of Energy, 1993; Valkenburg, 2008; European commission, 2003; Obernberger and Thek, 2004; Vassilev, 2010; Ahling, 1979; UNEP, 2005). Therefore, all types of AF, including the NRPP used at Balcones, can potentially substitute fossil fuels in cement production.

5. Potential for increasing the U.S. WTE capacity using AF

The 2013 use of AF in the cement industry effectively increased the total waste-to-energy (WTE) capacity of the U.S. from 27.4 million to 28.5 million tonnes, i.e. by 3.9%. If all energy for cement production in the US (310.5 billion MJ) was provided by EF similar to these used at Balcones facility (17.5 MJ/kg of NRPP), then about 17.7 million tonnes of the NRPP fuel would be needed. Therefore, the increase in the WTE capacity of 2013 or avoidance to landfill would be about 64.5%.

To quantify this benefit, it will be assumed that the current recovery from U.S. households of 33 million tonnes of paper fiber and 2.3 million tonnes of plastic wastes (Dolly, 2014) will be decreased by 17.7 million tonnes of NRPP, to be used in U.S. cement plants, similarly to the use of NRPP at the Balcones cement plant. This option will be compared with two alternatives: Landfilling the NRPP, as is done presently, and building additional WTE capacity for 17.7 million tonnes of MSW.

It is assumed that the NRPP collected and used in cement plants will consist of two thirds paper fiber (i.e. 11.8 million tonnes) and one third plastic wastes (5.9 million tonnes). Landfilling of 10 tonnes of MSW requires 1 m² of land (Dolly, 2014). Therefore, the use of the assumed amount of NRPP in the cement industry is associated with the conservation of 1.7 million square meters or 420.1 acres of land surface, plus the avoidance of about 53 million tonnes of CO_{2-eq} (2.9 tonnes CO_{2-eq} per tonne of NRPP).

For the comparison with WTE since the current capital cost of WTE power plants is between \$600 and \$800/tonne of MSW processed, the 2014 use of 17.7 million tonnes of NRPP in the U.S. cement industry would avoid the investment of \$10.6 to \$14.2 billion in new WTE capacity. However, the combustion of 17.7 million tonnes of MSW in WTE facilities would produce about 8.9 GWh of electricity and about 35 GWh of district heating/cooling or low-pressure steam for industrial uses (Zhang, 2013). Also, with WTE the recovery of about 10 wt% of ferrous and non-ferrous metals from the bottom ashes is possible. Metals are typically

with the highest value and demand in the market, which can go up to \$700 for non-ferrous aluminum. However, WTE is linked with the hazardous disposal of the fly ash produced, which will be at a small amount (0.17–0.89 million tonnes at an assumed 1 to 5 wt% of fly ash produced from the combustion of MSW), but the cost of processing and disposal is often up to \$50 to \$100 per tonne of fly ash. Although the production of NRPP requires some investment in mechanical processing, at MRFs and feeding systems at cement plants, it is evident that increasing the use of waste-derived fuels in cement production is economically more attractive than adding WTE capacity (Energy Recovery Council, 2016; Advancing Sustainable Materials Management, 2013). The cost of collection, transport and processing the NRPP at MRF facilities would be covered by the avoidance of landfill gate fee. Regarding energy benefits, the use of NRPP in cement production would avoid the mining of about the same tonnage of coal.

6. Conclusions

Cement production is one of the largest solids processing high-temperature process worldwide and it is associated with the emission of high amounts of CO_{2-eq} during production. The expanded use of AF in cement kilns will reduce the use of fossil fuels and the amount of wastes that need to be landfilled. This study has shown that the combustion of AF in the cement kiln system is being applied widely and offers definite environmental and energy advantages. The major findings of this study were:

1. The total energy consumption of the US cement industry is calculated at 4.2 MJ/kg of clinker for the year 2014 and is slightly reduced as compared to 1993, which was about 4.4 MJ/kg of clinker. The use of AF (tires, RDF, wood, liquid wastes, etc.) in the cement industry increased from 8.7% in 1993 to 18.9% in 2014 of the total energy consumption. Much higher levels have been reached in Germany (62%) and the Netherlands (83%). The use of "other solid wastes" in the U.S. cement industry increased from 90 thousand tonnes in 1993 to 1.3 million tonnes in 2014 which, at an assumed LHV of 18 MJ/kg, represented about 39.4% of the AF energy used in cement production (7.4% of the total fuel used).
2. On the assumption that the NRPP used at Balcones (17.5 MJ/kg) were to be used throughout the U.S. cement industry, the increase in the U.S. WTE capacity, and corresponding reduction in landfilling, would be about 17.7 million tonnes of NRPP, i.e. a 64.5% increase of the current U.S. WTE capacity.
3. The use of about 17.7 million tonnes of NRPP in the cement industry would be associated with the conservation of 1.7 million square meters or 420 acres of land surface, plus the avoidance of

about 53 million tonnes of CO_{2-eq}. Increasing the capacity of the U.S. WTE industry by 18 million tonnes would require a capital investment of \$11 to \$14 billion. Although the production of NRPP will require some investment in mechanical processing, at MRFs, and feeding systems at cement plants, it is evident that use of waste-derived fuels in cement production is economically more attractive than adding WTE plant capacity.

- The dioxin emissions examined in this study were an order of magnitude lower than the U.S. and E.U. standard (0.1 ng TEQ/Nm³ of stack gas). In a separate EEC study (Dwyer and Themelis, 2015), an inventory was created of all controlled (i.e. industrial) and non-controlled (i.e. landfill fires, back yard burning, etc.) sources of dioxins in the U.S. The results showed that the annual dioxin emissions of the entire U.S. cement industry amounted to 18 g TEQ, corresponding to 0.5% of all U.S. sources of toxic dioxins.
- The carbon dioxide emissions of four fuel scenarios were calculated using standard LCA methodology; they ranged from 100% high quality coal to 25% coal-75% engineered fuel. The results showed that the major GHG impact was the CO₂ generated during combustion of the fuel. The GHG benefit of using EF amounted to 3 tonnes of CO₂ per tonne of EF used in place of coal, when the avoidance of landfill gas emissions was included. The GHG emission is reduced from 390 kg CO₂ per tonne clinker for 100% coal to 137 kg for 75% EF and 25% coal.
- The overall conclusion from this study was that the use of MSW-derived fuels in cement production has no adverse impact on the cement production process or the quality of the product.

Acknowledgments

The authors would like to acknowledge the contribution of the Balcones plant personnel and the provision of operating data by Balcones engineers. The authors are also very grateful to Mr. Jose B. Martinez Madero of CEMEX for engaging the Earth Engineering Centers of Columbia University and City College of New York in this study.

Notation

AF	Alternative fuel
APC	Air pollution control
ASR	Automobile shredder residue
EF	Engineered fuel: shredded non-recycled plastics and paper residue (NRPP)
GHG	Greenhouse gases, expressed in CO _{2-eq}
LCA	Life cycle analysis
LHV	Lower heating value
MRF	Materials recovery facility
MSW	Municipal solid waste
NRPP	non-recycled plastics and paper residue (NRPP). Processed AF, called EF
PM	Particulate matter
RDF	Refuse derived fuels
SNCR	Selective non-catalytic reactor
TDF	Tire derived fuels
WTE	Waste-to-energy

References

Advancing Sustainable Materials Management, 2013. Fact Sheet Assessing Trends in Material Generation, Recycling and Disposal in the United States June 2015. https://www.epa.gov/sites/production/files/2015-09/documents/2013_advncng_smm_fs.pdf.

Ahling B., "Destruction of chlorinated hydrocarbons in a cement kiln", *Environ. Sci. Technol.*, 11/1979, ISSN: 0013-936X, Volume 13, Issue 11, p. 1377.

Ahn, S.Y., Eom, S.Y., Rhie, Y.H., Sung, Y.M., Moon, C.E., Choi, G.M., Kim, D.J., 2013. Application of refuse fuels in a direct carbon fuel cell system. *Energy* 51, 447–456.

Aho, M., Ferrer, E., 2005. Importance of coal ash composition in protecting the boiler against chlorine deposition during combustion of chlorine-rich biomass. *Fuel* 84, 201–212.

Ayllon, M., et al., 2006. Influence of temperature and heating rate on the fixed bed pyrolysis of meat and bone meal. *Chem. Eng. J.* 121, 85–96.

Borgianni, C., et al., 2002. Gasification process of waste containing PVC. *Fuel* 81, 1827–1833.

Brady, L.L., Hatch, J.R., 1997. Chemical analyses of middle and upper pennsylvanian coals from southeaster Kansas; current Research in Earth sciences, Kansas geological survey. *Bulletin* 240, 43–62.

Bye, G.C., Jan. 2011. Portland Cement, third ed. Thomas Telford Limited, London.

Chen, C., Habert, G., Bouzidi, Y., Jullien, A., 2010. Environmental impact of cement production: detail of the processes and cement plant variability evaluation. *J. Clean. Prod.* 18, 478–485.

CIWMB, 1992. California Integrated Waste Management Board. Tires as a Fuel Supplement: Feasibility Study, Sacramento, CA.

Cleary, J., 2009. Life cycle assessments of municipal solid waste management systems: a comparative analysis of selected peer-reviewed literature. *Environ. Int.* 35 (8), 1256–1266.

Conesa, J.A., et al., 2004. Complete study of the pyrolysis and gasification of scrap tires in a pilot plant reactor. *Environ. Sci. Technol.* 38, 3189–3194.

Dai, X., et al., 2001. Pyrolysis of waste tires in a circulating fluidized-bed reactor. *Energy* 26, 385–399.

De Queiroz Lamas, D., Fortes Palau, J.C., Rubens de Camargo, J., 2013. Renewable materials co-processing in cement industry: ecological efficiency of waste reuse. *Renew. Sustain. Energy Rev.* 19, 200–207.

Dept of Energy, report Chlorine in Coal and its Relationship with Boiler Corrosion, Technical report, September 1–November 30, 1993 United States.

US department of energy's (DOE's) Argonne national laboratory website. http://web.anl.gov/PCS/acsfuel/preprint%20archive/Files/44_1_ANAHEIM_03-99_0080.pdf.

Dolly, Shin, January 2014. Generation and Disposition of Municipal Solid Waste (MSW) in the United States—A National Survey. MS Thesis. EEE Columbia University. Available at: http://www.seas.columbia.edu/earth/wtert/sofos/Dolly_Shin_Thesis.pdf.

Dwyer, H., Themelis, N., 2015. Inventory of U.S. 2012 dioxin emissions to atmosphere. *Waste Manag.* 46.

Energy Recovery Council, 2016. Directory. <http://energyrecoverycouncil.org/wp-content/uploads/2016/05/ERC-2016-directory.pdf>. (Accessed December 2016).

European commission, July 2003. -directorate General Environment, Refuse Derived Fuel, Current Practice and Perspectives (B4-3040/2000/306517/MAR/E3).

Gallardo, A., Carlos, M., Bovea, M.D., Colomer, F.J., Albarrán, F., 2014. Analysis of refuse-derived fuel from the municipal solid waste reject fraction and its compliance with quality standards. *J. Clean. Prod.* 83, 118–125.

Galvez-Martos, J.L., Schoenberger, H., 2014. An analysis of the use of life cycle assessment for waste co-incineration in cement kilns. *Resour. Conserv. Recycl.* 86, 118–131.

Garg, A., Smith, R., Hill, D., Simms, N., Pollard, S., 2007. Wastes as co-fuels: the policy framework for solid recovered fuel (SRF) in Europe, with UK implications. *Environ. Sci. Technol.* 41 (14), 4868–4874.

Genon, G., Brizio, E., 2008. Perspectives and limits for cement kilns as a destination for RDF. *Waste Manag.* 28, 2375–2385.

Grammelis, P., et al., 2009. Pyrolysis kinetics and combustion characteristics of waste recovered fuels. *Fuel* 88, 195–205.

Gursel, A.P., Masanet, E., Horvath, A., Stadel, A., 2014. Life-cycle inventory analysis of concrete production: a critical review. *Cement Concr. Compos.* 51, 38–48.

Hassan, E.M., Steele, P.H., Ingram, L., 2009. Characterization of fast pyrolysis bio-oils produced from pretreated pine wood. *Appl. Biochem. Biotechnol.* 154, 182–192.

Huntzinger, D.N., Eatmon, T.D., 2009. A life-cycle assessment of Portland cement manufacturing: comparing the traditional process with alternative technologies. *J. Clean. Prod.* 17, 668–675.

IPCC, 2006. Guidelines Vol. II Energy. Table 1.2.

Jannelli, E., Minutillo, M., 2007. Simulation of the flue gas cleaning system of an RDF incineration power plant. *Waste Manag.* 27, 684–690.

Kaantee, U., et al., 2004. Cement manufacturing using alternative fuels and the advantages of process modelling. *Fuel Process. Technol.* 85, 293–301.

Kajaste, R., Hurme, M., 20 January 2016. Cement industry greenhouse gas emissions – management options and abatement cost. *J. Clean. Prod.* 112, 4041–4052. Part 5.

Kara, M., 2012. Environmental and economic advantages associated with the use of RDF in cement kilns. *Resour. Conserv. Recycl.* 68, 21–28.

Karstensen, H.H., 2008. Formation, release and control of dioxins in cement kilns. *Chemosphere* 70, 543–560.

Li, C., Nie, Z., Cui, S., Gong, X., Wang, Z., Meng, X., 2014. The life cycle inventory study of cement manufacture in China. *J. Clean. Prod.* 72, 204–211.

Littlefield, B.L., 2009. Characterization of Pecan Shells for Value-added Applications. Msc Thesis. department of Biosystems Engineering, Auburn University.

Liu, G.-Q., et al., 2001. Fundamental study of the behavior of chlorine during combustion of single RDF. *Waste Manag.* 21, 427–433.

Marceau, Medgar L., 2006. PCA, "Life Cycle Inventory of Portland, Cement Manufacture".

Mokrzycki, E., Uliasz-Bocheńczyk, A., Sarna, M., 2003. Use of alternative fuels in the

- Polish cement industry. *Appl. Energy* 74, 101–111.
- Montejo, C., Costa, C., Ramos, P., 2011. Analysis and Comparison of Municipal Solid Waste and Reject Fraction as Fuels for Incineration Plants, *Applied Thermal Engineering*.
- Nasrullah, M., Vainikka, P., Hannula, J., Hurme, M., Kärki, J., 2015. Mass, energy and material balances of SRF production process. Part 3: solid recovered fuel produced from municipal solid waste. *Waste Manag. Res.* 33 (2), 146–156.
- Committee on Coal Research Technology and Resource Assessments to Inform Energy Policy, 2007. National Research Council, National Academies Press.
- National Energy Technology Laboratory (NETL): Quality guidelines for system studies-Detailed Coal specification, 2012. DOE/NETL-2010/012111.
- Obernberger, Ingwald, Thek, Gerold, 2004. Physical characterization and chemical composition of densified biomass fuels with regard to their combustion behavior. *Biomass Bioenergy* 27, 653–669.
- Patel, C., Lettieri, P., Germanà, A., 2012. Techno-economic performance analysis and environmental impact assessment of small to medium scale SRF combustion plants for energy production in the UK. *Process. Saf. Environ.* 90 (3), 255–262.
- Piao, G., et al., 2000. Combustion test of refuse-derived fuel in a fluidized bed. *Waste Manag.* 20, 443–447.
- Rada, E.C., Ragazzi, M., 2014. Selective collection as a pretreatment for indirect solid recovered fuel generation. *Waste Manag.* 34 (2), 291–297.
- Rovira, J., Mari, M., Nadal, M., Schuhmacher, M., Domingo, J.L., 2010. Partial replacement of fossil fuel in a cement plant: risk assessment for the population living in the neighborhood. *Sci. Total Environ.* 408, 5372–5380.
- Rubber manufacturers association website, accessed August, 2016. <http://www.energyjustice.net/files/tires/files/scrapchn.html#anchor135840>.
- Scala, F., Chirone, R., 2004. Fluidized bed combustion of alternative solid fuels. *Exp. Therm. Fluid Sci.* 28, 691–699.
- Schneider, M., Romer, M., Tschudin, M., Bolio, H., 2011. Sustainable cement production. *Cement Concr. Res.* 41, 642–650.
- Senneca, O., Chirone, R., Salatino, P., 2002. A thermogravimetric study of nonfossil solid fuels. 2. Oxidative pyrolysis and char combustion. *Energy Fuels* 16, 661–668.
- Shih, P., Chang, J., Lu, H., Chiang, L., 2005. Reuse of heavy metal-containing sludges in cement production. *Cement Concr. Res.* 35 (11), 2110–2115.
- Skodras, G., et al., 2006. Pyrolysis and combustion characteristics of biomass and waste-derived feedstock. *Ind. Eng. Res.* 45, 3791–3799.
- Strazza, C., Del Borghi, A., Gallo, M., Del Borghi, M., 2011. Resource productivity enhancement as means for promoting cleaner production: analysis of co-incineration in cement plants through a life cycle approach. *J. Clean. Prod.* 19, 1615–1621.
- Trent, V.A., et al., 1982. Chemical Analyses and Physical Properties of 12 Coal Samples from the Pocahontas Field, Tazewell County, Virginia, and McDowell County, West Virginia; Geological Survey Bulletin 1528. United States Government Printing office, Washington.
- UNEP Chemicals, 2005. Standardized Toolkit for Identification and Quantification of Dioxin and Furan Releases. United Nations Environment Programme Chemicals, Geneva, Switzerland, p. 194.
- United Nations, 2010. Department of Economic and Social Affairs. *Energy Statistics Yearbook*.
- US EPA, 2008. Trends in Beneficial Use of Alternative Fuels and Raw Materials, Cement Sector, Draft.
- USGS, mineral Yearbook, Annual 1993–2016, Cement (last Accessed: 08/2016): <http://minerals.usgs.gov/minerals/pubs/commodity/cement/>.
- Valderama, C., Granados, R., Cortina, J.L., Gasol, C.M., Guillem, M., 2012. Implementation of best available techniques in cement manufacturing: a life-cycle assessment study. *J. Clean. Prod.* 25, 60–67.
- Valkenburg, C., Dec 2008. In: *Municipal Solid Waste to Liquid Fuels Synthesis*, vol. 1. US department of energy.
- Vassilev, Stanislav V., 2010. An overview of the chemical composition of biomass. *Nov 2009 Fuel* 913–933.
- VDZ, 1996. Concrete – hard as rock, strong on performance, fair to the environment. In: e.V. V.D.Z. (Ed.), *Environmental Compatibility of Cement and Concrete. Manufacture, Application and Use of Alternative Materials. Information from the German Cement Industry*, Germany.
- Wan, H.-P., et al., 2008. Emissions during co-firing of RDF-5 with bituminous coal, paper sludge and waste tires in a commercial circulating fluidized bed co-generation boiler. *Fuel* 87, 761–767.
- Wang, Z., et al., 2004. Laboratory investigation of the products of the incomplete combustion of waste plastics and techniques for their minimization. *Ind. Eng. Chem. Res.* 43, 2873–2886.
- WBCSD, 2006. Formation and Release of POPs in the Cement Industry. Cement Sustainability Initiative, World Business Council for Sustainable Development, p. 2006.
- Zhang, J., January 20, 2013. *Energy, Environmental and Greenhouse Gas Effects of Using Alternative Fuels in Cement Production*. MS- Thesis. Earth Engineering Centre, Columbia University, New York.
- Zhou, C., Zhang, Q., Arnold, L., Yang, W., Blasiak, W., 2013. A study of the pyrolysis behaviors of pelletized recovered municipal solid waste fuels. *Appl. Energy* 107, 173–182.
- Zou, J.H., et al., 2007. Modeling reaction kinetics of petroleum coke gasification with CO₂. *Chem. Eng. Process* 46, 630–636.