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Chlorine in waste-derived solid recovered fuel (SRF), co-combusted in cement kilns: A systematic review of sources, reactions, fate and implications

Spyridoula Gerassimidou[a], Costas A. Velis[a,*], Paul Williams[b], Marco J. Castaldi[c], Leon Black[a], Dimitrios Komilis[d]

[a] School of Civil Engineering, University of Leeds, Leeds, LS2 9JT, United Kingdom

[b] School of Chimical and Process Engineering, University of Leeds, Leeds, LS2 9JT, United Kingdom

[c] Chemical Engineering Department, City College, City University of New York, New York, NY 10031, United States

[d] Department of Environmental Engineering, Democritus University of Thrace, Xanthi, Greece

*Corresponding author: c.velis@leeds.ac.uk; Telephone: +44 (0) 113 3432327; Room 304, School of Civil Engineering, University of Leeds, Leeds, LS2 9JT, United Kingdom

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Abstract

Solid recovered fuel (SRF), a partly biogenic form of waste-derived fuel, can be used for replacing fossil fuels in cement kilns. Higher SRF uptake is limited mainly by its chlorine (Cl) content. Here we present a systematic literature review (PRISMA methodology) on the challenges induced by Cl during SRF co-combustion in cement kilns. We show that mean Cl content in average commercially manufactured SRF is at 0.76 w/w_d (± 0.14% w/w_d, 95% confidence). Cl is widely dispersed amongst various chemical compounds present in several waste items – not just PVC. The variability around mean Cl in SRF is at 36.7% (CV), notably lower than that in MSW, indicating effective variability reduction because of mechanical processing of MSW into SRF. During co-combustion, most Cl remains within the kiln system causing operating problems and only a small percentage (0.7 - 13%) is incorporated in the clinker, making it stickier. A significant proportion is captured by the air pollution control system. State of the art engineering practices can result in suitable SRF quality assurance, largely preventing Cl-related problems in the kiln. Further investigation on the impact of fuel feeding systems, on effect of Cl speciation and on controlling the sources of Cl variability are needed to improve confidence in SRF uptake.

Keywords: Chlorine; Solid recovered fuel; Cement kiln; Alternative raw material and fuel; Waste-to-energy; Circular Economy

Nomenclature

Chemical compounds and elements

Al	Aluminium
C_2Cl_4 (TCE)	Tetrachloroethene
C_2H_3Cl (VCM)	Chloroethene
$C_2H_4Cl_2$ (EDC)	Dichloroethane
C_2S	Belite
C_3H_5ClO (EPI)	2-(chloromethyl)oxirane
C_3S	Alite
Ca	Calcium
$Ca(OH)_2$	Calcium hydroxide
$Ca_2(SiO_4)_3(Cl)_2$	Chlorellestadite
Ca_2SO_4	Anhydrous calcium sulphate or anhydrite
$CaCl_2$	Calcium chloride
CaO	Calcium oxide (free lime)
CH_2Cl_2 (DCM)	Dichloromethane
Cl	Chlorine
Cl_2	Molecular chlorine
$COCl_2$ (CG)	Carbonyl dichloride (Phosgene)
Fe	Iron
$Fe(OH)_3$	Ferric hydroxide
Fe_2O_3	Ferric oxide or hematite
$FeCl_2$	Ferrous chloride
$FeCl_3$	Ferric chloride
H_2O	Water
HCl	Hydrochloric acid
Hg	Mercury
$K_2Ca_2(SO_4)_3$	Calcium-langbeinite
K_2O	Potassium oxide
KCl	Potassium chloride
MCl_2	Metal chloride
$MgCl_2$	Magnesium chloride
MO	Metal oxide

Na_2O	Sodium oxide
Na_2SO_4	Disodium sulphate
NaCl	Sodium chloride
NaClO	Sodium hypochlorite
NaOH	Sodium hydroxide
NH_4Cl	Ammonium chloride
O_2	Molecular oxygen
Pb	Lead
PCDDs	Polychlorinated dibenzo-p-dioxins
PCDFs	Polychlorinated dibenzo-p-furans
SiO_2	Dioxosilane (silica)
SO_2	Sulphur dioxide
SO_3	Sulphur trioxide
$TiCl_3$	Trichlorotitanium
Materials	
ABS	3,3-diphenyl-1-propanamine
CKD	Cement kiln dust
CPE	Chlorinated polyethylene
HDPE	High-density polyethylene
IBA	Incinerator bottom ash
LDPE	Low-density polyethylene
MSW	Municipal solid waste
PBT	Polybutylene terephthalate
PET	Polyethylene terephthalate
PP	Polypropylene
PPVC	Plasticized polyvinylchloride
PS	Polystyrene
PVC	Polyvinyl chloride
PVdC	Polyvinylidene chloride
RDF	Refuse derived fuel
SRF	Solid recovered fuel
WDF	Waste derived fuel
General	

BAT	Best available techniques
BC	Bomb calorimetry
CEN	European committee for standardization
EA	Elemental analyser
EC	European Commission
EfW	Energy from waste
EIA	Environmental impact assessment
ERFO	European Recovered Fuel Organisation
EU	European Union
FTIR	Fourier-transform infrared spectroscopy
GHG	Greenhouse gas
IC	Ion chromatography
ICP	Inductively coupled plasma
ICP-SFMS	Inductively coupled plasma-sector field mass spectrometry
MBT/MT	Mechanical-biological / mechanical treatment plant
MCP	Material, component and product
NCV	Net calorific value
NIR	Near infra-red
SFC	Schoeniger flask combustion
ToS	Theory of sampling
WRAP	Waste & Resources Action Programme
Selected units	
Mt	Million metric tonnes
Total [Cl]	Total chlorine concentration
w/w	Weight fraction on a wet basis
w/w_d	Weight fraction on a dry basis

1. Introduction

Cement manufacture is an intensive process with regards to fuel, electricity consumption and greenhouse gas (GHG) emissions (MPA Concrete centre, 2017, Saveyn, Eder, Ramsay, et al., 2016). Hence, the cement industry faces important challenges in meeting circular economy and decarbonisation goals, such as those set within the European Union (EU) (Cembureau.eu, 2016, WSP and DNV GL, 2015). The European cement industry already meets a considerable part of its fuel demand with combustible waste to remain cost competitive and attain environmental benefits (Samolada and Zabaniotou, 2014, Sharabaroff, Bernard, Lemarchand, et al., 2017, Uson, López-Sabirón, Ferreira and Sastresa, 2013). For example, in 2015, the cement industry in the UK replaced 42% of its thermal energy needs with waste derived fuels (WDF), an alternative to primary fossil fuels (MPA Concrete centre, 2017). Use of WDF in cement kilns is favoured due to: i) operating conditions, such as alkaline environment, good mixture of gases and products and high combustion temperature and flue gas residence time (Beckmann, Pohl, Bernhardt and Gebauer, 2012, Mokrzycki and Uliasz-Bocheńczyk, 2003); and ii) the experience of cement industry in handling a range of waste derived fuels (Garg, Smith, Hill, et al., 2009). In addition, cement kilns that adopted best available techniques (BAT) to comply with regulatory requirements do not require major alterations in the existing kiln system configuration (Schorcht, Kourti, Scalet, Roudier and Sancho, 2013).

Co-combustion of solid recovered fuel (SRF) in cement kilns is proven and energy efficient (European Commission, 2017): an established waste-to-energy process already widely adopted in Europe (ERFO and Cembureau.eu, 2015, Theulen, 2015). SRF is typically manufactured from non-hazardous components of solid waste and its composition meets national and EU specifications for co-combustion applications (BS 15359, 2011, Velis, Longhurst, Drew, Smith and Pollard, 2010). The thermal substitution rate of traditional fossil fuels by SRF in an individual plant currently can range between 40% and 70% (Saveyn, Eder, Ramsay, et al., 2016). For example, the Polish cement industry covered more than 50% of its fuel demand with SRF (ERFO, 2017); with the largest operator, HeidelbergCement, reaching a mass substitution rate of 70% in 2016 (ERFO, 2017).

SRF use in energy intensive industries contributes markedly to the reduction of GHG emissions due to its partial carbon neutrality, and abates acidification phenomena, because of lower sulfur content in comparison to fossil fuels (Garg, Smith, Hill, Simms and Pollard, 2007, Manninen, Peltola and Ruuskanen, 1997). Despite the energy consumed during SRF production, SRF co-combustion has high efficiency (current average factor ca 75% for cement kilns, defined as the heat recovered from waste input energy content (Saveyn, Eder, Ramsay, et al., 2016) leading to a positive overall balance (ERFO, 2019, Saveyn, Eder, Ramsay, et al., 2016).

SRF application in cement kilns and similar processes (e.g. lime kilns) can be regarded as an integrated part of modern waste management that substitutes fossil fuels and primary raw materials, improving resource efficiency (Hinkel, Blume, Hinchliffe, Mutz and Hengevoss, 2019). The European Commission (EC) policy recommendation on the use of waste-to-energy processes considers this option as one of the most energy efficient and compatible with circular economy (European Commission, 2017). Data reported by the European Recovered Fuel Organisation (ERFO) estimate the potential market to SRF at ca 50 million metrics tonnes (Mt) per year (Cuperus, 2019). Currently only at 13.5 Mt of SRF is used in Europe, of which 5 Mt is in the cement industry, with the potential to increase to close to 12 Mt (Cuperus, 2019, ERFO and Cembureau.eu, 2015). However, the market price of petcoke and the fees of alternative waste treatment options affect the financial attractiveness of SRF use in cement kilns (Hinkel, Blume, Hinchliffe, Mutz and Hengevoss, 2019). Different costs and revenues arise from SRF co-combustion depending on the waste management activities at municipal level: savings from sorting out of recyclables and substitution of fossil fuels, but there are costs related to collection, transportation, and operation of processes employed in mechanical- biological or mechanical treatment (MBT/MT) plants for SRF production (Hinkel, Blume, Hinchliffe, Mutz and Hengevoss, 2019, WRAP, 2009). Therefore, SRF use can be financially attractive for cement off-takers, if the cost reduction from the substitution of fossil fuels leads to an increased competitiveness and the associated risks are reduced (Hinkel, Blume, Hinchliffe, Mutz and Hengevoss, 2019). This condition can be achieved by improving the quality of SRF, which is subject to technical and quality assurance limitations, the most important of which is the chlorine (Cl) content (Cuperus, 2011, Lechtenberg and Diller, 2012).

Chlorine is a critical quality parameter for SRF, affecting technological performance in the cement production process (Cuperus, 2011, Lechtenberg and Diller, 2012) (e.g. cleaning costs due to build-up formation (Schorcht, Kourti, Scalet, Roudier and Sancho, 2013)). A high-quality SRF, with constant composition and low Cl content, is a prerequisite for increased substitution rates of fossil fuels by SRF in cement plants (Pomberger and Sarc, 2014). This can be achieved, to a certain degree, by using well selected waste feedstock and suitable sorting technologies employed in MBT/MT, such as near infra-red (NIR), to remove Cl containing compounds (Cuperus, 2011, Lechtenberg and Diller, 2012). To date, relevant research has focused on generic quality assurance during SRF production (Velis, Longhurst, Drew, Smith and Pollard, 2010), Cl in SRF produced in mechanical-biological or mechanical treatment (MBT/MT) plants (Velis, Wagland, Longhurst, et al., 2012), review of Cl fate in incineration, gasification, and pyrolysis (Lu, Huang, Bourtsalas, et al., 2018) and in generic use of alternative raw materials and fuels in cement kilns (Uson, López-Sabirón, Ferreira and Sastresa, 2013). But, despite its commercial importance, the role of Cl stemming from SRF is still poorly understood.

Here, we provide a systematic critical review of SRF application in modern cement kilns (cyclone preheater/precalciner kilns), focusing on Cl behaviour. Sources and fate of Cl in SRF during co-combustion in cement kilns is explicitly considered, aimed at defining key influential factors for the operational challenges induced by Cl presence in SRF. The two key research questions addressed were: (RQ1) sources and quantification of total Cl content in SRF; and, (RQ2) the impact of the Cl on cement production processes, when co-combusting SRF.

2. Methodology

2.1 PRISMA systematic review

In this critical review, we assembled and compared the accurately known information, with a view to provide useful guidance to cement kilns considering SRF usage. A systematic literature review was conducted following the PRISMA approach (Siddaway, 2014), consisting of five key stages: scoping, planning, searching, screening and eligibility (**Figure 1**). The PRISMA approach ensures the empirical evidence has been systematically collated and critically analysed to address the two key research questions (**Section 1**). Document types used in the scoping stage included: peer reviewed publications, industry reports, books, and PhD/ MSc theses. In the PRISMA planning stage, the two key research questions were split into specific sub-questions and search terms were selected covering materials, processing and operational conditions, as well as their combinations (**Table 1)**. Scientific databases (Scopus, Web of Science and Google Scholar) were searched for the keywords. The topic relevance of studies was assessed during the screening PRISMA stage, and the assessment covered document title, abstract and conclusions.

Exclusion criteria were posed during the eligibility stage for both key research questions. Regarding RQ1, only studies that described adequately the analytical method for Cl determination, the sample preparation and sampling process of SRF were included. Studies that did not clearly define whether the Cl was reported on wet or dry basis, and/or carried out sampling process deemed as insufficient (e.g. grab sampling) were excluded. For RQ2, limited exclusion criteria were applied, due to the confined literature. From the 743 documents identified as potentially relevant, only 176 were finally selected (ca 24%). Such exclusion criteria enabled existing actual research gaps to be identified, where reliable information was not available.

A plethora of studies investigate the behaviour of Cl in SRF under combustion conditions, focusing on energy from waste (EfW) plants and incinerators. Whereas not directly within the scope of cement kilns, such information was included in a separate section, because of the relevance of main thermochemical process (combustion), and for comparison purposes. The PRISMA flow diagram (**Figure 1**) presents the flow of information through the different phases of the systematic review, as applied here.

2.2 Data handling and statistics

Datasets on total Cl concentration referred to as Total [Cl] in this paper, in SRF, MSW and its constituting components were collected, classified based on the type of waste, and statistically analysed. Datasets were collected reporting the Total [Cl] from 9, 12 and 11 studies reporting the Total [Cl] as measured in MSW, SRF and different waste component categories, respectively.

Synthetic SRF samples (i.e. artificially constructed from individual components) have been used in past research to obtain insights into the fate of Cl in combustion processes (Cuperus, Van Dijk and De Boer, 2005, Wagland, Kilgallon, Coveney, et al., 2011, Wang, Huang, Li, et al., 2002, Wu, Glarborg, Frandsen, et al., 2011, Yasuhara, Katami, Okuda, Ohno and Shibamoto, 2001), but were excluded here from the statistical analysis. The composition of synthetic SRF samples is not standardized (e.g. an SRF component called ‘hard plastics’, may in fact contain considerably different combination of polymers in different studies), whereas NaCl and PVC were the exclusive or prevalent sources of Cl. However, in commercially produced SRF, the Cl load may also stem from other major sources, such as synthetic rubber and food additives, which questions the accuracy of such simulations.

The partitioning and terminology of waste components was inconsistent across studies. For example, the generic category of ‘plastics’ in certain cases was split further into more specific categories, such as ‘packaging’ and ‘non-packaging’ or ‘soft’ and ‘hard’ plastics. Specific categories were grouped here into more generic groups for consistency and comparability, using just five prevalent waste fractions, namely ‘plastics’, ‘paper/cardboard’, ‘wood’, ‘textiles’ and ‘rubber-leather’. Adjusted concentrations were obtained by multiplying the Total [Cl] in the specific item category with their proportion in the generic item category. The summation of these calculations gave us the Total [Cl] of the generic item category (‘plastics’). However, Total [Cl] can be expressed using different units: so normalization was performed to convert all estimates into % weight on a dry basis (% w/w_d) to enable comparisons.

Basic descriptive statistics and graphic analyses were performed in TIBCO StatisticaTM 13.3.0 software. The central location for the three sample populations (MSW, SRF and waste components categories) was determined (arithmetic mean and median) along with upper and lower 95% confidence intervals. Extent of relative variability was expressed as coefficient of variation (CV). We assumed that all the selected cases (studies that measured [Cl]) were of equal importance, although the number of samples was different in each case, and these were reported where available.

3. Chlorine content as determinant of SRF quality

The marketability of SRF as co-fuel requires quality assurance and control (Velis, Longhurst, Drew, Smith and Pollard, 2010). The European Committee Standardization (CEN) classified SRF quality based on three performance indicators with respect to economics (calorific content), technology (Total [Cl]) and environment (mercury content) (**Table 2**) (BS 15359, 2011). An overview of Cl limit values in SRF based on existing quality standards is reported by Velis, Longhurst, Drew, Smith and Pollard (2010) and Gendebien, Leavens, Blackmore, et al. (2003).

Chlorine, one of the key parameters used for quality assessment (Nasrullah, Vainikka, Hannula, Hurme and Oinas, 2016, Velis, Wagland, Longhurst, et al., 2012, Wu, Glarborg, Frandsen, et al., 2011), is limiting factor for SRF co-combustion in cement kilns (Pomberger and Sarc, 2014). The acceptable Cl levels depend on the configuration of the cement plant, with a typical range reported between 0.5-2% w/w (Schorcht, Kourti, Scalet, Roudier and Sancho, 2013). However, the recommended Total [Cl] in SRF, as applied by the cement industry, is less than 1% w/w_d, due to the tolerance of most thermal treatment processes in operation (Ibbetson and Wengenroth, 2007, Spurek, 2012, Velis, 2010, WRAP, 2012).

4. Total [Cl] uncertainty due to sampling and analytical determination

The origin (type of waste) of SRF makes the fuel highly heterogeneous. This may result in significant uncertainty of the ability of an SRF sample to represent the SRF from which it was drawn (known as ‘lot’). For the quality characterization of commercially produced SRF, samples are typically collected from the lot in an MBT/MT plant and delegated to the laboratory for analysis. Sub-sampling (a repetition of sampling process) then takes place to collect a small quantity of SRF that is submitted for analysis, defined as a ‘test portion’ (Prichard and Barwick, 2007), from the initial sample delivered by the MBT/MT plant. It is expected that the test portion would sufficiently represent the SRF lot that can reach up to 1,500 t (BS 15442, 2011).

Adherence to a correct sampling protocol is crucial to ensure representativeness during sampling (in the production plant) and sub-sampling (in the laboratory); but still, the measured [Cl] may vary considerably between samples due to high constitutional heterogeneity. For example, Spurek (2012) determined the Total [Cl] in individual SRF samples delivered to a Polish cement facility: in 12 out of 70 samples, the Total [Cl] exceeded the recommended value.

A comprehensive sampling plan in an MBT/MT plant is specified by CEN (BS 15442, 2011) and general guidelines for correct sub-sampling practices and equipment in the stage of laboratory analysis is given by Gerlach and Nocerino (2003). Based on the Gy’s theory (often referred to as ‘Theory of Sampling’: ToS), the sampling error is inversely proportional to sample mass and proportional to the cube of particle size of the sample (Pitard, 1993).

Thus, shredding decreases the sampling error and homogenizes the material, due to the creation of more uniform and equal probabilities of all particles in the material being included in the sample (Gerlach and Nocerino, 2003). Also, shredding helps the liberation of Cl in SRF, which otherwise might be occluded in large particles impeding its detection through analysis (Gerlach and Nocerino, 2003). The target particle size of an SRF test portion should be ≤1mm for [Cl] determination (BS 15408, 2011), but Cuperus, Van Dijk and De Boer (2005) suggested a particle size ≤0.5 mm for 100% Cl recovery.

During analytical determination, several methods are available for the quantification of [Cl]. This include: bomb calorimetry (BC) combined with ion chromatography (IC); quartz furnace methods; non-destructive methods, such as X-ray fluorescence; acid-destructive methods, such as inductive coupled plasma (ICP) technology; automated systems for elemental analysis (Ma and Rotter, 2008); and the Eschka method that can be applied directly in solid fuels (Rahim, Gao and Wu, 2014). The test portion varies over the range 0.05 - 100g depending on the analytical method.

Ma, Hoffmann, Schirmer, Chen and Rotter (2010) compared five analytical methods for [Cl] determination BC, Schoeniger flask method (SFC), elemental analyzer (EA), elution test and muffle furnace by measuring the [Cl] on 8 fractions of residual household waste. The SFC method illustrated higher Total [Cl] compared with other methods, while BC presented less [Cl] due to potential incomplete combustion in the bomb or Cl loss during bomb ventilation. In addition, Österlund, Rodushkin, Ylinenjärvi and Baxter (2009) concluded that a sample preparation method based on sintering, followed by inductively coupled plasma-sector field mass spectrometry (ICP-SFMS) analysis could be suitable in plastics and waste mixtures for [Cl] determination.

An extended literature review showed a variety of analytical methods for Cl determination in MSW and SRF (**Section 6 - Tables 4, 5**), while data availability is sparse. The most prevalent method was the BC-IC, the European standardized method for determination of Total [Cl] in SRF (BS 15408, 2011); yet other technologies, such as ICP and SFC, are widely used, as well. Further research is required to investigate the impact of sampling process on the uncertainty of representativeness of [Cl] in SRF samples and assess the influence of analytical methods with Round Robin test.

Despite the standard offline measurements conducted by laboratory analysis, there are online monitoring systems of HCl emissions produced during combustion, widely applied in cement plants or EfW boilers. These continuous monitoring systems are based on Fourier transform infrared (FTIR), gas filter correction, tunable diode laser and cavity ringdown technology (ICAC, 2013). Online measurements can offer sufficient control of HCl emissions during combustion and give a better understanding of measures to minimize these emissions by improving the operating conditions. In addition, online methods for monitoring of other chlorinated compounds in flue gas, such as dioxins (PCDDs) and their precursors, have been investigated (Gullett, Oudejans, Tabor, Touati and Ryan, 2011, Kuribayashi, Yamakoshi, Danno, et al., 2005, Liu, Jiang, Hou, et al., 2016, Nakui, Koyama, Takakura and Watanabe, 2011).

For a uniform kiln operation, the main properties of waste-derived fuels have to be monitored on a continuous basis (Bolwerk, 2004). So far, continuous monitoring systems are only available of chlorinated compounds in the flue gas (e.g. at the stack of the kilns). However, an online measurement system of Total [Cl] in the input waste based on Eschka method synchronized with the moving conveyor belt or the alternative transport system before burning would lead to considerably better SRF quality assurance.

5. A Cl categorization – where [Cl] load comes from?

The [Cl] in SRF can be differentiated based on two criteria. First, chemical speciation of Cl, which can largely be grouped as Cl chemically bound in *organic* or *inorganic* compounds; this split can often be coupled with the material, component and product (MCP) waste category, assuming just one prevalent form of chemical compound per MCP category. Second, in the literature the fate of Cl in the solid and gaseous phase during thermal treatment has also been used to classify it as '*incombustible'* or '*combustible'*. We detail and consider the relevance of these two categories below.

5.1 Chlorine categorization based on chemical compound/ material of origin

Chlorine can be identified as *organic* and *inorganic*, depending on its chemical bonding, which often correlates with the type of waste material (origin). The main source of *organic* Cl in SRF is plastics, whereas *inorganic* Cl derives from food waste: e.g. sodium chloride (NaCl) (common table salt) (Guo, Yang, Li, et al., 2001, Ma, Hoffmann, Schirmer, Chen and Rotter, 2010).

The characterization of Cl as *water soluble* and *water insoluble* may often indicate the presence of *inorganic* and *organic*, respectively (Vainio, Yrjas, Zevenhoven, et al., 2013), although it might be misleading, as some *organic chlorides* in SRF might be moderately soluble in water (Domalski, Ledford, Bruce and Churney, 1986), such as epichlorohydrin (EPI) (Open Chemistry Database, 2019a) and dichloromethane (DCM) (Open Chemistry Database, 2019b).

A main contributor to [Cl] load in MSW and sub-fractions is PVC, with Total [Cl] nearly 46 – 56% w/w (ECN Phyllis, 2019): a therefore unwanted polymer in SRF leading to high HCl release during fuel combustion (Heikkinen, Hordijk, De Jong and Spliethoff, 2004, Wang, Huang, Li, et al., 2002). Ma and Rotter (2008) reported that 63-72% of the entire [Cl] load in MSW derived from plastics is due to a considerable proportion of PVC in the 'plastic' fraction. This is in agreement with Watanabe, Yamamoto, Sakai and Fukuyama (2004) suggesting that the contribution of plastics to the [Cl] load in MSW was 63% and with Riber, Petersen and Christensen (2009) reporting that the non-recyclable plastic fraction constituted 60% of [Cl] load in household waste. Similarly, Velis, Wagland, Longhurst, et al. (2012) stated that 'plastic film' and 'packaging plastic' contributed 62% of total [Cl] load in SRF and Burnley (2007) claimed that 48% Total [Cl] in MSW was stemming from 'plastics'.

However, numerous *organic* Cl-containing compounds, other than PVC, are also used in many consumer products, delivering specific functionality (**Table 3**). Nearly one third of industrially produced Cl each year is predefined for PVC production, whereas a comparable amount goes to the production of isocyanates and oxygenates, prevalent chemicals in the packaging and textile industry (Deans, Dimas and Velis, 2016). So despite the literature having been preoccupied with identifying/removing the presence of PVC in SRF, a considerable load can be ascribed to other sources, either of lower [Cl] or in less prevalently used materials.

For example, PVdC has a higher [Cl] than PVC (nearly 71-73% w/w (Bhaskar, Negoro, Muto and Sakata, 2006, Zhao, Li, Zhang, et al., 2018)) and is typically used in packaging products; often combined with more affordable plastics, such as polypropylene (PP) for higher strength and moisture resistance (Marsh and Bugusu, 2007). Carbonyl dichloride (phosgene) is extensively used in the production of polycarbonate, which is used for glass replacements in items such as food containers, water and sterilisable baby bottles (Marsh and Bugusu, 2007), and isocyanates, which are precursors for the production of polyurethanes (Twitchett, 1974). Polyurethanes are present in various consumer products, such as furniture, clothing, shoes, electrical devices, sealants and adhesives (ECI, 2019, Krone, Ely, Klingner and Rando, 2003), eventually becoming MSW.

Organic Cl can also be present in MSW in the form of epichlorohydrin, a compound used in the production of elastomers in rubber production, making synthetic rubber a significant potential contributor of [Cl] load, as verified by previous researchers (Nasrullah, Vainikka, Hannula, Hurme and Oinas, 2016, Velis, 2010, Velis, Wagland, Longhurst, et al., 2012). Velis, Wagland, Longhurst, et al. (2012) investigated the properties of components in commercially produced SRF and identified that almost 14.5% of total [Cl] load in SRF was attributed to ‘shoes’, concluding that synthetic rubber within shoes constitute a highly chlorinated waste component and should therefore be separated out from the SRF. The Total [Cl] of shoes was found in a range of 3.6-6% w/w_d (Rotter, Kost, Winkler and Bilitewski, 2004, Velis, 2010).

Epichlorohydrin (EPI) is also used for the manufacture of epoxl resins, which are mainly used in packaging and paper manufacture (Santodonato, Lande, Howard, Orzel and Bogy, 1980), in insulation of electronic equipment, in adhesives and fibre-reinforced composites (Barton, 1985). For that reason, SRF item components, categorized as 'adhesives', 'furniture' and 'cables', are also considered as potential high sources of Cl. For example, Velis (2010) measured the Total [Cl] of 'cables', considered as electric wires with variable plastic casing, close to 2.5% w/w_d.

Yet other sources of *organic* Cl, occurring less frequently in MSW, contribute to the overall [Cl] load. For example, chlorinated polyethylene (CPE) is a thermoplastic elastomer produced by the chlorination of polyethylene (PE) with advanced properties (ViaChemical, 2017), finding its way typically into mixed unsorted or residual MSW. Dichloromethane (DCM) and tetrachloroethene (TCE) are chlorinated solvents with various applications, as well (**Table 3**).

Inorganic Cl, consisting of alkali metal chlorides such as NaCl, KCl, $CaCl_2$ (**Table 3**), is prevalent in the 'food waste' fraction. The high proportion of 'food waste' in MSW makes NaCl the major contributor to the [Cl] load in MSW (Guo, Yang, Li, et al., 2001, Ma, Hoffmann, Schirmer, Chen and Rotter, 2010, Wu, Wang, Chai, Takahashi and Shimaoka, 2016). But, *inorganic* Cl is not entirely present only in 'food waste'. Ziegler-Natta catalysts used in the polymerization of polyolefin are composed of $TiCl_3$ and $MgCl_2$ leading to the presence of Cl in polyofelin plastic (Shamiri, Chakrabarti, Jahan, et al., 2014). Additionally, packaging components, such as plastic bottles of chlorinated detergents, may often contain remnants of chloride compounds such as sodium hypochlorite - the most common chloride used for bleaching (Zoller, 2008).

Chlorine dispersion in various waste components restricts the ability to reduce the [Cl] in SRF below a limit depending on waste composition (Rotter, Kost, Winkler and Bilitewski, 2004). But, the technological performance of SRF can be improved by increasing the mass fraction of waste components that contain negligible amounts of Cl such as ‘paper/cardboard’, ‘wood’ and ‘non-PVC plastics’ (Nasrullah, Vainikka, Hannula, Hurme and Oinas, 2016). ‘Paper’ is a main component in MSW with low [Cl] contributing considerably to reducing the overall [Cl] load in SRF (Velis, 2010). The low [Cl] in paper is due to residuals of chlorine dioxide and hypochlorite may remain in paper after the bleaching process (Bajpai, 2014).

Nevertheless, such components can be contaminated with chlorides due to the absorption of water percolated through food waste in the residual waste stream (cross-contamination) (Uchida, Kamo and Kubota, 1988), e.g. in a high compaction collection vehicle, which is a prevalent collection means for residual MSW in developed countries (Edjabou, Jensen, Götze, et al., 2015). Domalski, Ledford, Bruce and Churney (1986) reported that in MSW where ca. 45% w/w was ‘paper’, 65-80% of the entire *inorganic* [Cl] load could be allocated to ‘paper’. Cross-contamination is a partly unavoidable condition, which can decrease the market value of SRF or even make waste fractions unsuitable for further processing (Smith, Cheeseman and Blakey, 2009). Cross-contamination of SRF from residuals might occur at different stages of waste processing- not only during waste collection. Despite the beneficial role of shredding in waste homogenization (**Section 4**), over-pulverization may lead to considerable cross-contamination and, therefore, should be avoided (Velis, 2010). The level of cross-contamination can decrease if the performance of sorting unit operations in an MBT/MT plant, which can be evaluated by the purity, recovery rate and yield factor, is high (Riddell, Gardner, Perez-Gonzalez, Lopes and Martinez, 2015, Velis, 2010).

5.2 Chlorine categorization based on its fate (partitioning) in EfW combustion

The fate of Cl in thermal processing has been more extensively studied for EfW plants than cement kilns. Hence, we considered the relevant literature to obtain insights on the combustion behaviour of Cl and its species. Based on the fate of Cl in thermal processing, the proportion of Cl converted into HCl in the flue gas during combustion, could be considered to be a ‘*combustible*’ fraction, while the proportion that remains in incinerator bottom ash (IBA) considered as ‘*incombustible*’ (Watanabe, Yamamoto, Sakai and Fukuyama, 2004). Such terminology is based on Cl fate within a particular system – that of EfW combustion plants (incineration plants), practically defined as the partitioning coefficients, calculated as part of material flow analysis, widely applied for metals (e.g. (Brunner and Monch, 1986)), often interpreted in relation to their volatility (e.g. volatile vs. lithophilic elements or compounds). An association between Cl abundance and metal partitioning behaviour has been reported (Saqib and Bäckström, 2014), with the presence of Cl decreasing the transfer of trace elements to bottom ash by forming highly volatile metal chlorides. Specifically, elevated Cl levels led to an increased distribution of highly (Hg, Cd) and moderately (Pb, Zn, As) volatile chlorides to fly ash due to the affinity between trace elements and HCl released during combustion. This is also confirmed by earlier studies (Astrup, Riber and Pedersen, 2011, Belevi and Moench, 2000).

Investigations on the fate of Cl during waste combustion in incinerators (older MSW combustion plants without energy recovery) and EfW boilers indicated that HCl is an important product (Kanters, Van Nispen, Louw and Mulder, 1996, Uchida, Kamo and Kubota, 1988, Wang, Huang, Li, et al., 2002). MSW combustion in a fluidized bed reactor at high temperature (900°C) resulted in ca. 45% w/w of Cl load partitioning to the gaseous phase and 55% w/w to the solid phase (Liu, Pan and Riley, 2000). Chang and Huang (2002) investigated the Cl speciation and material flow in two MSW incinerators with different air pollution control equipment: the first was equipped with cyclone, followed by a dry lime scrubbing system with a fabric filter; and the second was equipped with an electrostatic precipitator followed by a wet scrubber. They found that ca. 60% w/w of the total Cl input at both incinerators was captured by the air pollution control devices, while the remaining discharged into IBA.

Research into the correlation between the *organic* vs. *inorganic* bonding of Cl in compounds and its fate in thermal processing has been carried out. Ma and Rotter (2008) proposed that *organic* and *inorganic* Cl may present different thermal behavior. During combustion major products of *inorganic* Cl are HCl, Cl_2 and salt depositions, whereas *organic* Cl contributes more to the HCl release. This relative contribution is due to either incomplete decomposition of NaCl (*inorganic* Cl) leading to Cl deposition in IBA and/or slower release rates of Cl ions that might promote the potential formation of metal chlorides (Wang, Chiang, Lin, Tsai and Sun, 1999, Wang, Huang, Li, et al., 2002, Wey, Chen, Wu, Yu and Tsai, 2006). Wang, Huang, Li, et al. (2002) compared the amount of HCl released by NaCl compared to PVC, finding that the HCl conversion rate of total Cl input from PVC was between 60 and 65%, whereas the same conversion rate from NaCl was in a range of 30-40%. A possible reason was that PVC contains hydrogen which reacts with Cl forming HCl, while NaCl reacts with water vapour to produce HCl. Another consideration was that the affinity of Cl to sodium is higher than of Cl to hydrogen (Wey, Ou, Liu, et al., 2001).

Material flow analysis of Cl during MSW combustion showed that *organic* Cl contributes considerably to the formation of *combustible* Cl. Watanabe, Yamamoto, Sakai and Fukuyama (2004) reported 75% of *organic* Cl in input material resulted as *combustible* and 25% as *incombustible*. Almost 100% of Cl in PVC is irreversibly transformed into HCl and released at a temperature range of 200-400ºC, regardless of the operating conditions of the combustion process (e.g heating rates), making PVC a crucial precursor of HCl formation (Lu, Purushothama, Hyatt, et al., 1996, Ma and Rotter, 2008); (**Reaction 1**) (Kaufmann, 1997). Kanters, Van Nispen, Louw and Mulder (1996) combusted MSW with and without the PVC fraction and deduced that the removal of PVC materials from MSW could decrease HCl release by up to 40%.

$$\text{PVC: } (CH_2CHCl)_n \rightarrow HCl + nC_2H_2 \qquad (1)$$

However, Ma, Hoffmann, Schirmer, Chen and Rotter (2010) suggested that the correlation between *organic-inorganic* and *combustible-incombustible* Cl content is not robust, because Cl volatilization is affected, also, by the temperature profile of the reactor. This is in agreement with Wey, Chen, Wu, Yu and Tsai (2006), who investigated HCl formation during waste incineration in a fluidized bed reactor and showed that 63.5%, 98.7% and 87.5% of *organic* Cl was transformed into HCl at 600ºC, 700ºC and 800ºC, respectively.

Despite the inability to clearly associate Cl categorization into *combustible* and *incombustible* to physico-chemical properties of MCPs or to combustion processing conditions, this distinction may be of operational relevance for EfW plants. However, it may not be entirely suitable to extrapolate combustion results from EfW technological settings to cement plants with regards to the fate of Cl during combustion, because the processes, reactions and sinks are customarily more complicated in the latter, as we demonstrate in **Section 7**.

6. Chlorine content in SRF and its sources/ components

6.1 Chlorine content in MSW

Typical values of Total [Cl] in MSW reported in previous studies are presented in **Table 4**. The 95% confidence interval of Cl content in MSW was 1.00 ±0.35% w/w_d and the minimum and maximum values were 0.37% w/w_d and 1.94% w/w_d, respectively. This is in line with a report that summarized the best practices in the cement industry and reported that the Total

[Cl] in MSW ranged between 0.5-1.5% w/w (Sharabaroff, Bernard, Lemarchand, et al., 2017). Also, Table 4 presents the proportion of organic and inorganic Cl present in MSW, which was in the range of 50-75% and 50-25% of the Total [Cl], respectively. Data on the combustible and incombustible Cl was limited.

6.2 Chlorine content in SRF

Typical values of Total [Cl] in SRF are presented in **Table 5**. The 95% confidence interval was 0.76 ±0.14% w/w_d; with minimum and maximum values of 0.31% w/w_d and 1.38% w/w_d, respectively. The results are in agreement with Gendebien, Leavens, Blackmore, et al. (2003), reporting that a typical Total [Cl] of WDF from household waste is in the range 0.3-1% w/w. A wider range was given by Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen (2015) of 0.1 to 3.9% w/w_d. For a particular SRF, with a high PVC content, Total [Cl] was reported at >1% w/w_d (Sarc, Lorber, Pomberger, Rogetzer and Sipple, 2014).

It has been argued that the wide range of Total [Cl] in SRF can be largely attributed to the heterogeneity of solid waste. Deans, Dimas and Velis (2016) modelled the extent of uncertainty of [Cl] in SRF based on three different scenarios, depending on material composition, generating probability distributions using Monte Carlo simulations. They showed a greater spread of [Cl] in the case of SRF with high content in plastics.

6.3 Chlorine content in SRF components

Typical values of Total [Cl] in waste component categories of MSW and consequently SRF are shown in **Table 6**. 'Rubber-leather' has the highest average Cl content, indicating its potential role on [Cl] load in SRF. However, the category 'rubber-leather' covers a wide range of materials, and the limited number of observations give rise to inconsistent [Cl] values and prevent any further estimation. Of the waste component categories, 'plastics' had the highest Total [Cl] followed by ‘textiles’, ‘wood’ and ‘paper/cardboard’. Sørum and Task (2001), remind us that [Cl] may vary in some waste components depending on their physical properties or production methods. For example, [Cl] in paper depends on the bleaching process used for the removal of lignin, and [Cl] in plastics varies according to the polymer type.

The values of Total [Cl] in MSW, SRF and waste component categories derived from the literature review are depicted in **Figure 2**. The boxplots of SRF and MSW are of a similar scale, but the interquartile range in MSW is wider than in SRF. This indicates a higher variability, which is also confirmed by the relative variation (CV) (**Table 4-6**). The relative variation of SRF is more than 30% lower than MSW. This significant variability reduction indicates the effectiveness of sorting and shredding unit operations employed in MBT/MT plants on SRF homogenization.

Regarding the waste component categories, the highest spread of [Cl] values was observed in ‘plastics’, while the lowest was in ‘paper/cardboard’ (**Figure 2**). However, their relative variation (**Table 6**) is almost equal (with a CV nearly 53% for ‘plastics’ and 52% for ‘paper/cardboard’) indicating similar relative Cl dispersion. The highest relative variation was found in ‘wood’ probably due to the extreme [Cl] value (0.98% w/w_d) presented in the boxplot, which increased CV from 56% to 88%. However, we cannot omit this value, since considerable variation in [Cl] can be present depending on tree species (Pettersen R.C, 1984) and cross-contamination of such materials with chlorides through water absorption. Second in the descending order of [Cl] variability comes ‘textiles’ (with a CV almost 65%). This might be related to the nature of this category, as ‘textiles’ include both synthetic and natural fabrics leading to a variable [Cl]. A further division of this component category into sub-fractions may give a better estimation, but research preoccupation with ‘textiles’ is not as great as with ‘plastics’. **Table 7** illustrates the Total [Cl] in sub-fractions of the ‘plastic’ category, as defined by previous studies. These values were used to construct the dataset of ‘plastics’ used in our statistical analysis, as explained in **Section 2.2.**

The variability of [Cl] is high for both waste mixtures (MSW and SRF) and waste components due to uncontrolled factors, such as seasonality and origin (Beckmann, Pohl, Bernhardt and Gebauer, 2012), and controlled factors such as sampling and analytical methods (Gerlach and Nocerino, 2003). However, the relative variation of [Cl] is higher for waste component categories than SRF. This was unexpected as SRF is a mixture of waste components while waste fractions have more uniform constitution. We hypothesize that the higher relative variability of [Cl] in waste component categories is attributed to the different sorting rules and name definitions applied by researchers during characterization of waste fractions. In contrast, SRF statistically analysed here was the outcome of commercial processes without any artificial sorting during the analysis stage.

7. SRF application in the cement industry

Modern cement plants are the main end-users of SRF (ERFO, 2019), but Cl is one of the key limiting factors (Cuperus, 2011, Lechtenberg and Diller, 2012, Pomberger and Sarc, 2014). Whereas for EfW boilers there is ample information on the implications and fate of Cl during combustion of solid waste (Albina and Themelis, 2005, Chen, Zhang, Ma, Rotter and Wang, 2015, Hilber, Maier, Scheffknecht, et al., 2007, Liu, Pan and Riley, 2000, Pettersson, Niklasson and Moradian, 2013), the situation has not been extensively studied in the cement industry. This is despite the preoccupation of the cement off-takers with total [Cl] in SRF. The main difference between the two combustion technologies is that cement plants operate at higher temperatures, with clinkering taking place at 1450ºC, and with temperatures of 2000ºC in the flame of the main burner and 900-1200ºC in the precalciner (Cement Sustainability

Initiative, 2005, Jensen, 1999). A typical bed temperature in EfW boilers fluctuates between 780-900ºC (Miller and Tillman, 2008).

Co-combustion of SRF in the cement industry is largely considered a sustainable waste recovery option, offering cost savings means by avoiding consumption of primary fossil fuels, decarbonising the cement industry and preventing wider natural resources (Chatziaras, Psomopoulos and Themelis, 2016, MPA Concrete centre, 2017, Uson, López-Sabirón, Ferreira and Sastresa, 2013). However, SRF utilization may affect the clinker manufacturing process and induce operating problems in the kiln, depending on the fuel quality. For example, SRF particles, which are larger than solid fossil fuels, can lower the gas temperature and degrade the cement quality, due to delayed ignition and slower thermal conversion, resulting in insufficient burnout and a large amount of particles falling onto the clinker bed (Liedmann, Wirtz, Scherer and Krüger, 2017).

The 'raw meal' used for the clinker production comprises primarily limestone and clays, providing a source of silicates and aluminates, plus some iron oxides (Fe_2O_3) (Schorcht, Kourti, Scalet, Roudier and Sancho, 2013). Limestone has a high absorption capacity of acidic gases (e.g. HCl) due to its alkalinity and high content of reactive calcium oxide (CaO), which has high affinity for Cl (Karstensen, 2006, Lanier, Stevens, Springsteen and Seeker, 1996). This may importantly push the [Cl] in the cement over the 0.1% w/w threshold stipulated in EN197-1:2000. For that reason, the output quality of clinker must be well monitored to detect any effects when SRF is used (Schorcht, Kourti, Scalet, Roudier and Sancho, 2013).

The substitution rate of traditional fossil fuels by using SRF in cement kilns depends on the feeding point (primary firing, secondary firing), the conveying technology, plus the fuel composition (Scur, 1998). There are a number of potential fuel feeding ports in a cement plant (Cement Sustainability Initiative, 2005, Sarc, Lorber, Pomberger, Rogetzer and Sipple, 2014, Schorcht, Kourti, Scalet, Roudier and Sancho, 2013). Feeding via the main burner at the rotary kiln outlet end requires high quality SRF. Low quality SRF can be added via secondary burners to the riser duct at the kiln inlet. At precalciner burners lower quality SRF can be used than in the main burner. Finally, addition via a feed chute to the precalciner requires better quality SRF than the precalciner. The SRF quality depends on the feedstock/waste composition and the technology/processes used for the SRF production (Nasrullah, Vainikka, Hannula and Hurme, 2013, Nasrullah, Vainikka, Hannula, Hurme and Kärki, 2015). Improving quality therefore may lead to higher cost (Sharabaroff, Bernard, Lemarchand, et al., 2017).

7.1 Fate of Cl in a modern cement kiln

Cl behavior in a cement kiln has been examined by the application of WDF with high [Cl] and by the addition of Cl in raw meal, since Cl is introduced to the kiln through the raw materials and fuel (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015, Enders and Haeseli, 2011, Saxena, 2003). Chlorine may either leave the system (‘sinks’) through the clinker, the baghouse dust and unburned chlorinated emissions in the stack or remain in the kiln (‘stock’). Chlorine hardly leaves the kiln via clinker in modern cement kilns, due to its high volatility and the operating conditions (Holderbank, 1992). Exceptions can be explained mainly by the cooling down of sintering zone, Cl encapsulation in big material lumps, and/or formation of high internal Cl cycles (Holderbank, 1992, Schorcht, Kourti, Scalet, Roudier and Sancho, 2013).

The only recent study investigating the fate of Cl during co-combustion of SRF in modern cement kilns, i.e. with a preheater/precalciner system, was by Zhan, Fu, Chen, et al. (2016). The effects of the bypass system on Cl circulation in modern kilns were examined by calculating the Cl balance. SRF was fed into the precalciner under a 6.5% mass substitution rate. The results showed that the absence of a bypass system induced 12.7% and 14.6% of input Cl to flow out through the clinker and flue gas respectively, while the remaining 72.7% remained in the inner surface of the preheater or accumulated in the cement ash. The presence of a bypass system with a bypass ratio close to 2%, led to 11.8%, 10.9% and 17.3% of the Cl input in the flue gas, the clinker and the bypass dust respectively, while 60% was the difference between input and output Cl.

Chlorine can be found in the clinker in the form of inorganic chlorides with the most prevalent being $CaCl_2$ (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015). Chloride compounds, such as chlorellestadite ($Ca_{10}(SiO_4)_3(SO_4)_3Cl_2$), HCl, Cl_2, alkaline earth chlorides (e.g $MgCl_2$, $CaCl_2$) and transition metal chlorides, can be found in the pre-heater zone (Saint-Jean, Jøns, Lundgaard and Hansen, 2005, Sidhu, Kasti, Edwards and Dellinger, 2001). Chlorellestadite ($Ca_{10}(SiO_4)_3Cl_2$) can be formed in the preheater due to the reaction of HCl gas with the hot meal, and is affected by the degree of contact between the kiln gases and the cement kiln dust (CKD) (Saint-Jean, Jøns, Lundgaard and Hansen, 2005). Chlorellestadite can also be found in build-ups and kiln rings; build-ups contain both organic and inorganic Cl compounds, with the most prevalent being chlorellestadite, KCl and NaCl (Bhatty, 2011). **Figure 3** depicts the fate of Cl and the most prevalent forms of Cl during SRF co-combustion in modern cement kilns, accompanied by a generalised mass flow of typical Cl distribution.

7.1.1 Chlorine circulation in a kiln-preheater system

In a modern cement kiln, Cl is volatilized and is returned with the flue gas back to the preheater, where it condenses and returns to the high temperature zone, creating a continuous volatilization-condensation reaction. This can cause instability in the system and increase the energy consumption (Enders and Haeseli, 2011). Chlorine volatilization takes place in the burning zone of the kiln at a temperature range of 900ºC to1200ºC (Enders and Haeseli, 2011), while condensation occurs in the riser pipes and in the lower cyclones of the preheater

(Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015). Thus, at temperatures between 700ºC and 900ºC, mixtures with low melting points are formed (Schorcht, Kourti, Scalet, Roudier and Sancho, 2013). The material and the hot-gas flows in the kiln are arranged in counterflow, causing Cl circulation (**Figure 4**), that can be controlled by reduced [Cl] in fuel and raw materials, and/or installation of a bypass system (Enders and Haeseli, 2011). Chlorine circulation is determined by the phase equilibrium between liquid and gaseous alkali metal chlorides (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015, Enders and Haeseli, 2011). Also, a high SO_3 to alkali ratio is crucial for Cl cycling/circulation, as sulfation of alkali chlorides may occur at temperatures above 900ºC (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015).

Chlorine vaporization can take place via two mechanisms. In the first mechanism inorganic chlorides are volatilized directly in the gas phase without intermediate reactions due to high temperatures, as follows (Enders and Haeseli, 2011):

$$KCl_{(s)} \leftrightarrow KCl_{(g)} \tag{2}$$

$$NaCl_{(s)} \leftrightarrow NaCl_{(g)} \tag{3}$$

Also, organic Cl can be volatilized as HCl (**reaction 1**) and/or alkali chlorides react with water vapours in the gas phase at temperatures above 1000ºC, forming HCl thus(Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015, Saint-Jean, Jøns, Lundgaard and Hansen, 2005):

$$NaCl_{(g)} + H_2O_{(g)} \leftrightarrow NaOH_{(g)} + 2HCl_{(g)} \tag{4}$$

In the second mechanism, HCl reacts with silicate minerals and melts forming alkali chlorides in the flue gas (Enders and Haeseli, 2011):

$$K_2O_{(s,m)} + 2HCl_{(g)} \leftrightarrow 2KCl_{(g)} + H_2O_{(g)} \tag{5}$$

$$Na_2O_{(s,m)} + 2HCl_{(g)} \leftrightarrow 2NaCl_{(g)} + H_2O_{(g)} \quad (6)$$

Condensation products are minerals derived from the reactions between alkali chlorides in gas phase and ingredients of the hot meal (Enders and Haeseli, 2011):

$$CaO_{(s)} + 4KCl_{(s)} + 6SO_2 + 3O_2 + 2H_2O \leftrightarrow K_2Ca_2(SO_4)_{3(s/l)} + 4HCl_{(g)} \quad (7)$$

$$CaO_{(s)} + 3SiO_2 + 2KCl_{(s)} + 4SO_2 + O_2 \leftrightarrow Ca_2(SiO_4)3(SO_4)_3(Cl)_{2(s/l)} + K_2SO_4 \quad (8)$$

7.1.2 Effects of Cl input on clinker quality

Chlorine primarily reacts with alkalis and the remainder (less than 1%) reacts with the free lime (CaO) contained in the raw meal, due to its stronger affinity with alkalis than calcium (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015, Holderbank, 1992, Sutou, Harada and Ueno, 1999). The remaining Cl is absorbed by the free lime forming $CaCl_{2(s/l)}$ (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015, Sutou, Harada and Ueno, 1999) (**reaction 9**). This has a low melting point (772ºC) such that, despite the low quantities, the raw meal becomes sticky (Vainikka, Enestam, Silvennoinen, et al., 2011). This phenomenon reduces the ability of raw meal to flow, causing operating problems in the system (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015, Karstensen, 2006).

$$CaO_{(s)} + 2HCl_{(g)} \leftrightarrow CaCl_{2(s/l)} + H_2O_{(g)} \quad (9)$$

Kwon, Kim, Chu, et al. (2005) investigated the effect of Cl on clinker quality by adding NH_4Cl to raw materials. Total [Cl] ranged from 0.5% to 2% w/w and the mineralogical composition of clinker was analyzed by X-ray diffraction. The results showed that Cl acted as an accelerator for the liquid-solid clinkerization reaction, forming $CaCl_{2(s/l)}$ by reducing the amount of free lime and alite (C_3S) and increasing the amount of belite (C_2S) in the clinker. While some belite is welcome because of its ease of grindability (Maki, Ito, Tanioka, Ohno and Fukuda, 1993), the destabilisation of alite is unwelcome since it is this phase, typically constituting 45-65% of the clinker, which makes the main contribution to strength, particularly at early ages (Talaber, 1982).

Based on a previous study, a mass substitution rate of 50% SRF in cement kilns may induce a fivefold increase in clinker chloride contents (Gendebien, Leavens, Blackmore, et al., 2003). Despite this, the chloride concentrations were still below typical limits for chloride in Portland cement, of 1000 mg Cl kg^{-1} (0.1% w/w) (BS 197-1, 2000). This was due to complete Cl vaporization in the sintering zone induced by the high vapor pressure (Gendebien, Leavens, Blackmore, et al., 2003, Lechtenberg and Diller, 2012).

Despite the impact on clinker quality, $CaCl_2$ formation promotes Cl circulation. It can react with oxygen and water vapours producing Cl_2 and HCl, as follows (Shigapov, Novoshilova, Vereshchagin, Anshits and Sokolovskii, 1988):

$$CaCl_{2(s/l)} + 0.5O_{2(g)} \rightarrow CaO_{(s)} + Cl_{2(g)} \qquad (10)$$

$$CaCl_{2(s/l)} + H_2O_{(g)} \rightarrow CaO_{(s)} + HCl_{(g)} \qquad (11)$$

In addition, $CaCl_2$ can form metal chlorides either with direct (**reaction 12)** or indirect chlorination (**reactions 13a and 13b**) at 1000°C (Chan and Kirk, 1999):

$$CaCl_{2(s/l)} + MO_{(g)} \rightarrow CaO_{(s)} + MCl_{2(g)} \qquad (12)$$

$$CaCl_{2(s/l)} + 0.5O_{2(g)} \rightarrow CaO_{(s)} + Cl_{2(g)} \qquad (13a)$$

$$Cl_{2(g)} + MO_{(g)} \rightarrow MCl_{2(g)} + 0.5O_{2(g)} \qquad (13b)$$

HCl can also be formed by the sulfation of $CaCl_2$ increasing the risk of corrosion of the steel wall (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015):

$$CaCl_{2(s/l)} + 0.5O_{2(g)} + SO_{2(g)} + H_2O_{(g)} \rightarrow CaSO_{4(g/l)} + 2HCl_{(g)} \qquad (14)$$

7.1.3 *Effects of Cl input on flue gas*

HCl is formed during combustion of SRF in cement kilns (with formation starting from 200ºC) (Lechtenberg and Diller, 2012), but the Total [Cl] in SRF is not critical regarding gaseous pollutants, due to the alkaline environment in the kiln (Genon and Brizio, 2008). Schorcht, Kourti, Scalet, Roudier and Sancho (2013) reported data from continuous dust measurements in the clean gas of 253 kilns and found that HCl emissions from European cement kilns ranged from 0.02 to 20 mg Nm^3, depending on the composition of raw materials and fuels, the age and design of the plant, and the requirements of the regulatory authority.

Gendebien, Leavens, Blackmore, et al. (2003) reported that SRF use in cement kilns under 50% mass substitution can lead to an almost 24% increase in HCl emissions, which can be captured by the alkaline scrubber effect (Karstensen, 2006), while a 15% mass substitution rate does not affect the HCl emissions (Rovira, Mari, Nadal, Schuhmacher and Domingo, 2010). This statement is in agreement with Fyffe, Breckel, Townsend and Webber (2012) who carried out burn tests of 130 t of SRF fed into a precalciner with a 10% energy substitution rate, finding no increase in HCl emission rates.

Despite HCl emissions, other chlorinated organic compounds, such as PCDDs/Fs, can be formed in the presence of Cl (Karstensen, 2008). However, modern preheated cement kilns that use non-hazardous wastes as co-fuels present PCDDs/Fs emissions lower than 0.1 ng l-TEQm^{-3} under well managed operation, indicating that waste can be used as co-fuels (Karstensen, 2008, Peters, Hughes and Mournighan, 1983, Schorcht, Kourti, Scalet, Roudier and Sancho, 2013). Genon and Brizio (2008) reported that SRF does not affect the emissions of chlorinated-micropollutants due to the thermal conditions and alkaline environments in the kiln.

7.1.4 *Effects of Cl input on cement kiln dust (CKD)*

According to the UK Environment Agency (2001), the majority of the Cl remains in the kiln, while a small part is precipitated with particulates in the de-dusting system. The volatile Cl is mainly concentrated in the fine particles of the CKD, produced by the exhaust gas cooling. The coarse dust particles are returned to the kiln (via injection into the raw feed or directly to the mill (Environment Agency, 2001)), while the fine particles, known also as Cl bypass dust, are treated as waste (Saito, Ueno, Harada, Okamura and Suzuki, 2011).

Specifically, heavy metals are vaporized and react with $CaCl_2$ forming unwanted metal chlorides (**reactions 12-13**) through the sintering process, which are concentrated within the CKD in the bag filter (Miura, Sato, Suzuki, Oogami and Yazawa, 2001, Zhang, Bogush, Wei, et al., 2018). The most prevalent chloride salts during SRF co-combustion are the strong hydration retarders $PbCl_2$ and $ZnCl_2$ (Vainikka, Bankiewicz, Frantsi, et al., 2011), since Pb is used as a metal stabilizer in plastics, textiles and leather and Zn as acid scavenger and filler in plastics (Zweifel, Maier and Schiller, 2009).

When increasing quantities of Cl bypass dust are produced, it becomes difficult to recover all of the Cl, necessitating the installation of a bypass system (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015, Enders and Haeseli, 2011, Sutou, Harada and Ueno, 1999). As a rule of thumb, the [Cl] input has to be less than 400 g tn^{-1} clinker, otherwise a bypass system is required (Lechtenberg and Diller, 2012). The system consists of a probe which extracts and cools the exhaust gas, a cyclone which separates and returns coarse dust to the kiln, and a bag filter or electrostatic precipitator which captures the bypass dust and the exhaust gas. Although the bypass system offers a technical solution for Cl removal, additional capital cost and energy consumption is required (Enders and Haeseli, 2011). Besides, the disposal of bypass dust is difficult due to environmental restrictions (Lechtenberg and Diller, 2012).

Sutou, Harada and Ueno (1999) reported that extraction of 2% and 5% of kiln exhaust gas (bypass ratio) could lead to nearly 50% and 90% Cl removal, respectively. A more recent study found that Cl removal from the system was nearly 18% with a 2% bypass ratio (Zhan, Fu, Chen, et al., 2016).

7.2 Operating problems induced by the presence of Cl in modern cement kilns

Chlorine-related problems in cement kilns are attributed to Cl circulation, CKD recycling and Cl absorption from free lime. The latter can lead to sticky particles (from the clinker and CKD) generating material build-ups on the wall (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015). Build-ups are formed on the preheater cyclones due to excessive Cl circulation (Holderbank, 1992) causing blockages (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015, Karstensen, 2006), and pressure drops in the preheater tower (Turnell, 2001). An insulating layer can be formed on the electrodes in the electrostatic precipitator reducing the electric field and, thus, lowering its capture efficiency, leading to increased dust emissions (Schorcht, Kourti, Scalet, Roudier and Sancho, 2013). Enders and Haeseli (2011) reported that [Cl] in hot meal can serve as an indicator to evaluate the risk of build-up formation, suggesting a [Cl] more than 3.5% w/w is detrimental. Successive deposition of particles and gas condensation may cause ring formation (Dominguez, Gómez-Millán, Alvarez, et al., 2010) leading to high cleaning costs and kiln shutdowns (Schorcht, Kourti, Scalet, Roudier and Sancho, 2013). Ring formation enhances Cl circulation since the available opening of the kiln for the gas and material flow decreases (Enders and Haeseli, 2011, Tran and Barham, 1991).

Corrosion potential in cement kilns is complicated due to multiple temperature zones, plus variable numbers of preheaters and raw meal composition (Potgieter, 1997). Low levels of oxygen in combination with high [Cl] input may cause corrosion of the kiln shell, resulting in thinning below the mechanical limits of kiln shell stability (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015, Enders and Haeseli, 2011). Yet evidence suggests that the corrosion mechanism in cement kilns is similar to those in steam boilers (Jons and Ostergard, 1999, Potgieter, Godoi and Van Grieken, 2004).

The corrosion mechanism can be described by a number of chain reactions: alkali chlorides react with metal oxides (**reaction 15a**) or HCl is oxidized (**reaction 15b**), forming Cl_2, which reacts with the iron metal shell (**reaction 16**). These products are oxidized and hydrolysed forming metal oxides and HCl (**reaction 17 and 18**) (Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen, 2015, Lee, Themelis and Castaldi, 2007, Sharobem and Castaldi, 2012):

$$2Na^{*}Cl_{(g)} + Fe_2O_3 + 0.5O_2 \rightarrow Na_2Fe_2O_4 + Cl_2 \qquad (15a)$$

$$2HCl_{(g)} + 1/2O_{2(g)} \leftrightarrow Cl_{2(g)} + H_2O_{(g)} \qquad (15b)$$

$$Fe_{(s)} + Cl_{2(g)} \rightarrow FeCl_{2(s)} \qquad (16)$$

$$12FeCl_{2(s)} + O_{2(g)} \rightarrow 8FeCl_{3(g/l)} + 2Fe_2O_{3(s)} \qquad (17)$$

$$FeCl_{3(s)} + 3H_2O_{(g/l)} \rightarrow 3HCl_{(g)} + Fe(OH)_{3(s/l)} \qquad (18)$$

* Sodium can be replaced by potassium

8. Mitigation of Cl-related problems in cement kilns

From a technical perspective, high SRF quality can be achieved by producing a homogeneous waste output with low Total [Cl]. However, the inherent heterogeneous composition of solid waste and current waste management practices make the production of homogeneous SRF for MBT/MT plants a major technical challenge (Velis, Longhurst, Drew, Smith and Pollard, 2010). SRF end-users need a suitable SRF quality assurance to deal with the inherent heterogeneous output material, which may lead to variable thermal behavior and, therefore, difficulty to adjust the thermal recovery processes (Beckmann, Pohl, Bernhardt and Gebauer, 2012). There are actions and processes that can improve SRF quality regarding Cl-related problems in the stage either of waste processing or co-combustion. **Table 8** summarises the main measures that can be taken for mitigating Cl-induced problems at each of these two stages.

In **Table 8**, we see that the measures have their merits and demerits, but it is not easy to address the best option. The effectiveness of each depends on the quality and quantity of input waste feedstock and the existing framework conditions at municipal level (Hinkel, Blume, Hinchliffe, Mutz and Hengevoss, 2019). A combination of these measures could bring synergistic effects, whereas quality assurance procedures are undoubtedly required. Maintaining constant operating conditions during thermal treatment and optimizing mechanical processes employed in MBT/MT plants could ensure SRF quality assurance.

9. Conclusions

We have investigated, by way of systematic review, the role of Cl in SRF during co-combustion in cement kilns. The findings offer insights into the actual and perceived challenges around Cl as the key limiting factor for wider use and higher uptake of SRF in such industrial applications. Specifically, we conclude that:

- Cl content in SRF is widely dispersed amongst various items present in MSW, and is present in many chemical forms. Specifically, we demonstrate here that various forms of organically bound Cl (e.g. PVdC, EPI, DCM, TCE and phosgene) and inorganically bound Cl (e.g. NaCl, KCl and $CaCl_2$) could constitute considerable sources due to their wide application in packaging, textile and food industry materials. This information could enable differentiation of sorting strategies during mechanical processing of MSW to produce an SRF which is low in Cl, going beyond the current preoccupation with identifying and/or removing only PVC. Statistics have confirmed results from individual studies in identifying the generic waste categories ‘rubber-leather’, ‘plastics’ and ‘textiles’ as considerable potential sources of Cl load. Inversely, ‘paper/cardboard’ and ‘wood’ were confirmed as low in Cl combustibles, and can thus be used as high fractions to lower the SRF Total [Cl] content.

- Industrial off-takers of SRF are mainly interested in reducing the variability around the mean for Cl, so that they can adjust their thermal recovery processes around a tightly defined, and therefore manageable, Cl level (e.g. install a Cl bypass or not, choose air pollution control equipment, and ensure statistical regulatory compliance). Evidence collated here demonstrates that the Total [Cl] variability (measured as coefficient of variation) around the average is ca. 30% lower for SRF in comparison with MSW. This finding exemplifies the effectiveness of mechanical processing equipment employed in MBTs/MTs plants in homogenizing the SRF output, despite the similar levels of Total [Cl] found in SRF and MSW. The relative variation of Total [Cl] was over 50% for MSW and waste components due to unavoidable (constitutional heterogeneity), partly unavoidable (cross-contamination) and controlled factors such as sampling, sub-sampling and analytical methods. This underpins the importance of research to quantify the opportunities for manipulating the controlled factors to further reduce the variability of Cl in SRF.
- Despite Cl being considered the main technical limiting factor for SRF application in the cement industry, our systematic review confirms that the related phenomena have not been extensively studied. Most of the evidence exists for cases of high-Cl streams and not for MSW-derived SRF (e.g. hazardous waste or highly chlorinated substances

present in the raw meal). Cl circulation may affect the clinker manufacturing process and cause operating problems in the kiln, such as build-up and ring formation, corrosion and clinker quality degradation. Most Cl remains in the system ('stock') in the form of alkali chlorides, a small percentage is incorporated in the clinker as $CaCl_{2(s/l)}$ making the clinker sticky. A significant proportion is captured by the baghouse in air pollution control dust. SRF co-combustion does not play a critical role in the emission of gaseous pollutants due to the alkaline environment and thermal conditions prevailing in cement kilns.

- Chlorine-related problems during SRF co-combustion in cement kilns can be controlled, and a number of options are available. Keeping the substitution rate at conservative levels is effective, yet not cost efficient. Well-monitored operating conditions are necessary. Installation of a bypass system if effective, yet costly, energy consuming and the bypass dust introduces an additional major solid waste liability. Finally, SRF of a suitable quality may be used, which depends on the production stage (waste processing). The recommended Total [Cl] in SRF, as applied in fact by the cement industry, is nearly 1% w/w_d, but it depends on the requirements and configuration of the cement plant (with a typical range reported of 0.5-2% w/w). Further investigation into the impact of feeding point of SRF in cement kilns on the fate of Cl and the resultant implications is required.
- Finally, we clarify here that the distinction of Cl into *combustible* and *incombustible* may be of operational relevance for EfW plants, but may not be suitable to describe the fate of Cl during combustion within cement plants. The speciation of relevance is only that of organically bound and inorganic compounds (salts), which result in different chemical reaction pathways.

Our investigation into the sources, reactions and fate of Cl in cements kilns using SRF suggests that despite the major knowledge gaps, there is sufficient but currently underused information that could potentially allow advanced design of SRF. By adopting better practices SRF producers could lower the average Cl concentration and control its variability around the mean, enabling higher substitution rates and widespread application.

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Table 1. Search terms and two main research questions systematically reviewed, according to PRISMA principles.

Search terms			
Materials	**Compounds**	**Application**	**Fate of Cl**
Solid recovered fuel Waste derived fuel Municipal solid waste Household waste Alternative fuel	Chlorine Chloride(s) Hydrogen chloride Chlorinated compounds	Cement plant Cement kiln Suspension preheater Precalciner kilns Combustion	Clinker Flue gas Cement kiln dust Corrosion Operating problems

Main research questions (RQ1, RQ2), and specific sub-questions	
RQ1: Quantification of chlorine (Cl) content in SRF	**RQ2: How Cl presence in SRF affects cement production process**
Are there quality standards of SRF for Cl content?	How is the Cl content distributed during co-combustion in a cement kiln system?
What is the total Cl concentration of SRF and its components?	What are the main chloride compounds that can be found during combustion of SRF in a cement kiln?
What are the main sources of Cl in SRF?	Does the feeding port of SRF affect the Cl behaviour during combustion in a cement kiln?
Do the measured values accurately represent the total Cl concentration in SRF?	How does Cl affect the clinker quality?
Are there parameters that may affect the measurement of Cl concentration?	How does Cl affect the flue gas emissions?
What are the main forms of chloride compounds that can be found in SRF?	How does Cl affect the operation of the kiln?
What are the possible categorisation/ speciation of Cl (e.g. chlorinated compounds) in SRF?	What is the limit of Cl content for SRF utilization in cement plants?

Table 2. Rapid quality characterisation ('Classification') of SRF according to EN 15359:2011(BS 15359, 2011), the core document of the most comprehensive set of standards available for SRF. Total Cl content, measured by specific compliance statistics and sampling process, is one of the key 3 indicators of quality.

Classification characteristic	**Performance category**	**Statistical measure***	**Unit**	**Classes**				
				1	**2**	**3**	**4**	**5**
Net calorific value (NCV)	Economic	Arithmetic Mean	MJ kg^{-1} (ar)	≥ 25	≥ 20	≥ 15	≥ 10	≥ 3
Chlorine (Cl) [Total]	Technological	Arithmetic Mean	% (d)	≤ 0.2	≤ 0.6	≤ 1.0	≤ 1.5	≤ 3
Mercury (Hg)	Environmental	Median	mg MJ^{-1} (ar)	≤ 0.02	≤ 0.03	≤ 0.08	≤ 0.15	≤ 0.50
		80th percentile	mg MJ^{-1} (ar)	≤ 0.04	≤ 0.06	≤ 0.16	≤ 0.30	≤ 1.00

* Quality control practices require statistical tools to ensure the consistency and compliance to current specifications of SRF properties, such as unbiased estimates for central tendency (arithmetic mean, median), spread of values (e.g. standard deviation, coefficient of variation), and upper/lower confidence limits. Many studies reported SRF properties as individual numbers without including the uncertainty, leading to misleading impression. The properties of SRF need to be specified by a level of uncertainty (Velis, 2010).

Table 3. Main compound forms and MCPs applications of *organic* and *inorganic* Cl in SRF.

Cl type	**Name**	**Symbol**	**Formula**$^{(1)}$	**Uses**	**Ref.**
Organic	Ethylene dichloride	EDC	$C_2H_4Cl_2$	VCM production	(Kurtz, 1975)
	Chloroethene	VCM	C_2H_3Cl	PVC production	(Summers, 2000)
	Polyvinyl chloride	PVC	$(C_2H_3Cl)_n$	Tubes, pipes, films, toys, flooring, and other construction production; packaging industry; textiles	(Summers, 2000)
	Polyvinylidene chloride	PVdC	$(C_2H_2Cl_2)_n$	Packaging of cheese, tea, coffee, poultry and meat products, catsup and salad dressing bottles; textiles	(Marsh and Bugusu, 2007)
	Epichlorohydrin	EPI	C_3H_5ClO	Textiles and rubber (elastomer production); coating of food and beverage cans, plastics, cables, adhesives and fibre-reinforced composites (epoxy resin production); paper products (tissues, towelling, and beverage filters)	(Barton, 1985, Santodonato, Lande, Howard, Orzel and Bogy, 1980)
	Chlorinated Polyethylene	CPE	$(C_5H_9Cl)n^{*}$	Cable jacketing, rubber, roofing, automotive and industrial hose and tubing, moulding, base polymer	(ViaChemical, 2017)
	Dichloromethane	DCM	CH_2Cl_2	Paint removal; textiles, shoes, adhesives, cables and furniture (polyurethane foam production); decaf coffee	(Belay, Ture, Redi and Asfaw, 2008, Jimoda, 2011, Kaefferlein, Ruehl and Bruening, 2011)
	Tetrachloroethene	TCE	C_2Cl_4	Textiles; cleansers; adhesives and waterproof materials	(Hickman, 2000, US Department of Health and Human Services, 2019a)

Cl type	Name	Symbol	Formula(1)	Uses	Ref.
	Carbonyl dichloride (Phosgene)	CG	$COCl_2$	Textiles, shoes, adhesives, cables, furniture (polyurethane foam production); glass container and bottles	(Marsh and Bugusu, 2007, Twitchett, 1974)
Inorganic	Sodium hypochlorite		NaClO	Most common chloride bleach	(Zoller, 2008)
	Sodium chloride		NaCl	Table salt	(Guo, Yang, Li, et al., 2001, Ma, Hoffmann, Schirmer, Chen and Rotter, 2010, Themelis, 2010)
	Titanium (III) chloride Magnesium chloride		$TiCl_3$ $MgCl_2$	Ziegler-Natta catalysts for PP and PE polymerization	(Shamiri, Chakrabarti, Jahan, et al., 2014)
	Potassium chloride		KCl	Food processing (flavouring agent, nutrient supplement, pH control agent, stabilizer and thickener)	(Van Buren, Dötsch-Klerk, Seewi and Newson, 2016)
	Calcium chloride		$CaCl_2$	Food additives (canned vegetables and fruits, cheese, beverages and ice cream)	(Keowmaneechai and McClements, 2002, US Department of Health and Human Services, 2019b)

[*] (Manaila, Stelescu and Craciun, 2012) , (1) (ChemSpider)

Table 4. Dataset of typical values of Cl in MSW reported in previous studies.

Type	**Total [Cl] (% w/w_d)**	**Inor. [Cl] (% of Total Cl)[1]**	**Or. [Cl] (% of Total Cl)[2]**	**Com. [Cl] (% of Total Cl)[3]**	**Incom. [Cl] (% of Total Cl)[4]**	**Analytical method**	**No. samples[5]**	**Ref.**
MSW[d]	0.46 0.89	24.4 24.2	75.6 75.8			ASTM E775-81	15	(Domalski, Ledford, Bruce and Churney, 1986)
MSW	0.46	50.0	50.0	82.3	17.7	Quartz combustion	n.d.[a]	(Kanters, Van Nispen, Louw and Mulder, 1996)
MSW	0.37					Solid waste incineration system	13	(Chang, Chen and Chang, 1998)
MSW	1.05					BC-IC	n.d.[a]	(Sørum and Task, 2001)
MSW	0.59			75.4	24.6	Quartz furnace - IC	n.d.[a,b]	(Watanabe, Yamamoto, Sakai and Fukuyama, 2004)
Household waste	0.85					ICP-SFMS	n.d.[a,b,c]	(Riber, Petersen and Christensen, 2009)
MSW[d]	1.27 1.63	33 21	67 79			Eschka method	4 3	(Prawisudha, Namioka and Yoshikawa, 2012)
MSW	1.50					ISO 10304-1:2009	n.d.[a]	(Nasrullah, Hurme, Oinas, Hannula and Vainikka, 2017)
MSW	1.94					EA	n.d.[a]	(Chen, Zhang, Ma, Rotter and Wang, 2015)
Arithmetic mean	**1.00 (0.65, 1.36)***							
CV (%)**	**52.76**							
d.f.***	**10**							

[1] Inorganic; [2] Organic; [3] Combustible; [4] Incombustible; [5] Number of independent measurements; * No. replicates per sample; [a] n.d.: not determined; [b] Calculated based on MSW composition and Cl values of hand-sorted MSW fractions; [c] Batches of 80-1200 tonnes of household waste from 3 different cities collected for the calculation of Cl; [d] for two different cities, Baltimore County, Maryland and New York, Brooklyn; [e] two different MSW samples; * Lower and upper 95% confidence interval; ** Coefficient of Variation; *** Degrees of freedom

Table 5. Dataset of typical values of Cl in SRF reported in previous studies.

Type	**Total [Cl] (% w/w_d)**	**Analytical method**	**Origin (type of waste)**	**No. samples**[1]	**Ref.**
SRF[2]	0.31	Incineration system	MSW	15	(Chang, Chen and Chang, 1998)
SRF$_{SBS}$[3]	0.57	Combustion -IC	MSW and commercial	n.d.[a]	(Hilber, Maier, Scheffknecht, et al., 2007)
SRF	0.71	BC-IC	MSW	45	(Velis, 2010)
SRF	0.41	Data from supplier	Commercial & industrial	n.d.[a]	(Vainikka, Bankiewicz, Frantsi, et al., 2011)
SRF	0.45	BC-IC	Commercial & industrial	2	(Vainikka, Enestam, Silvennoinen, et al., 2011)
SRF	0.69	BC-IC	Household and commercial	30	(Velis, Wagland, Longhurst, et al., 2012)
SRF	0.85	BC-IC	Household and industrial	10	(Aldrian, Sarc, Pomberger, Lorber and Sipple, 2016)
SRF$_{SBS}$[3]	0.49 0.89	BC-IC	MSW	n.d.[a]	(Glorius, 2014)
SRF[4]	1.38 1.08	BC-IC	Several streams	15 21	(Sarc, Lorber, Pomberger, Rogetzer and Sipple, 2014)
SRF	0.71 0.80	ISO 10304-1:2009	MSW, commercial & industrial	3 3	(Nasrullah, Hurme, Oinas, Hannula and Vainikka, 2017)
SRF[5]	0.91 0.69 1.19	BC-IC	MSW	3 9 6	(Edo-Alcón, Gallardo and Colomer-Mendoza, 2016)
SRF	0.92	Eschka method	MSW	70	(Spurek, 2012)
SRF	0.60	BC-IC	MSW	3	(Nasrullah, Vainikka, Hannula, Hurme and Oinas, 2016)
Arithmetic mean	**0.76 (0.62, 0.90)**[*]				
CV (%)[**]	**36.70**				
d.f[***]	**17**				

[1]Number of independent measurements; * No. replicates per sample; [2]It is reported as refuse derived fuel (RDF) similar to SRF without compliance with EU specification, due to oldness of the study; [3]SRF developed by the German organisation Remondis; [4]Different qualities of SRF provided by different suppliers; [5]MSW reject flows form 3 different MBT plants; [a] n.d.: not determined; [*] Lower and upper 95% confidence interval; [**] Coefficient of Variation; [***] Degrees of freedom;

Table 6. Dataset of typical values of Cl in waste component categories reported in previous studies.

	Plastics	Paper/ cardboard	Wood	Textiles	Rubber-leather	No. samples^1	Ref.
Total [Cl] (% w/w$_d$)	1.14	0.15	0.05	1.10	8.00	3	(Nasrullah, Vainikka, Hannula, Hurme and Oinas, 2016)
	1.84	0.50	0.10	0.40	2.20	3	(Rotter, Kost, Winkler and Bilitewski, 2004)
	0.54	0.27	0.40	0.32	0.36	n.d.^{a}	(Guo, Yang, Li, et al., 2001)
	3.62	0.41	0.39	1.19	7.00	n.d.^{a}	(Uchida, Kamo and Kubota, 1988)
	1.55	0.10	0.27	1.17	-	2	(Ma, Hoffmann, Schirmer, Chen and Rotter, 2010)
	2.40	0.18	0.23	0.41	-	4	(Watanabe, Yamamoto, Sakai and Fukuyama, 2004)
	3.88	0.30	-	-	-	n.d.^{a}	(Sørum and Task, 2001)
	2.62	0.24	0.22	0.22^b 0.23^b	0.76	18 ^{1} 2^{2,3,4,5}	(Chiemchaisri, Charnnok and Visvanathan, 2010)^1, (Velis, 2010)^{2,3,4,5}
		0.33^c 0.04^c	0.98			n.d.^{a}	(Heikkinen, Hordijk, De Jong and Spliethoff, 2004)
		0.25^d 0.21^d				15	(Domalski, Ledford, Bruce and Churney, 1986)
Arithmetic mean	**2.20 (1.23, 3.17)***	**0.25 (0.17, 0.33)***	**0.33 (0.09,0.57)***	**0.63 (0.26, 1.00)***	**3.66 (0.36, 8.00)$^{\#}$**		
CV (%)**	**52.94**	**51.59**	**87.83**	**64.75**	**97.86**		
d.f.***	**7**	**11**	**7**	**7**	**4**		

^{1,2,3,4,5} refer to Plastics, Paper/cardboard, Wood, Textiles and Rubber-leather, respectively; ^{a} n.d.: not determined; ^{b}each value corresponds to textiles/fabrics and carpets/mats, respectively; ^{c} each value corresponds to cardboard and glossy paper; ^{d} the values derive from two different cities; * Lower and upper limits of 95% confidence interval for sample mean; ** Coefficient of Variation; *** Degrees of freedom; ^{1} Number of independent measurements * No. replicates per sample; $^{\#}$ range between minimum and maximum used due to data inconsistency

Table 7. Overview of Cl content in specific plastic categories or polymers.

SRF component	**Cl (% w/w_d)**	**Reference**
Packaging plastics	1.00	(Rotter, Kost, Winkler and Bilitewski, 2004)
Non-packaging films	0.60	
Other plastic products	8.50	
ABS	0.04	(Heikkinen, Hordijk, De Jong and Spliethoff, 2004)
HDPE	0	
PBT	0	
PET	0	
PP	0	
PVC	51.31	
Paper & cardboard	0.10	(Ma, Hoffmann, Schirmer, Chen and Rotter, 2010)
Hard plastics (bottles and containers)	1.00	
Soft plastics (bags and soft films)	0.60	
Non-packaging plastics	6.15	
Plastic carrying bags	2.51	(Chiemchaisri, Charnnok and Visvanathan, 2010)
Other plastic bags	1.23	
Other plastics	3.95	
Durable plastic	0.02	(Velis, 2010)
Plastic film	1.28	
Other packaging	1.52	
Plastic (hard)	1.60	(Nasrullah, Vainikka, Hannula, Hurme and Oinas, 2016)
Plastic (soft)	0.83	

Table 8. Measures for mitigation of Cl-related problems in cement kilns and quality improvement of SRF in the stage of production and thermal treatment.

	Measures	**Description**	**Demerits**	**Reference**
SRF production (waste processing)	Near-infra-red (NIR)-sorting technology	Optical sorting technology that enables plastic materials to be separated by polymer type (used for sorting out the PVC fraction).	Difficulty to treat cost-efficiently dark plastic components and incapability to sort out *inorganic* Cl.	(Morrish, Morton, Myles and Wilkinson, 2010, Pomberger and Sarc, 2014, Sarc, Lorber, Pomberger, Rogetzer and Sipple, 2014, Velis, Wagland, Longhurst, et al., 2012)
	Selective collection	Pre-treatment of MSW for direct extraction of PVC-containing materials.	No clear evidence whether different types of selective collection are more efficient than mechanical processing in dedicated MBT/MT plants.	(Rada and Ragazzi, 2014)
	Addition of low Cl-containing materials	Increasing the mass fraction of paper and cardboard due to its biogenic content and lower [Cl].	Decrease of SRF calorific value and consequently its financial attractiveness.	(Velis, Longhurst, Drew, Smith and Pollard, 2010, Velis, Wagland, Longhurst, et al., 2012)
	Torrefaction	Conversion of SRF into a coal-like material (char) at low temperature (200 - 350°C) leading to the improvement of SRF quality. The fuel becomes more homogeneous and its calorific value can be increased from ca. 20 to 25 MJ/kg improving the SRF attractiveness as an energy source, whereas Total [Cl] can decrease by 35 to 55% due to Cl migration from feedstock to the gas phase.	No commercial torrefaction unit is available yet.	(Białowiec, Pulka, Stępień, Manczarski and Gołaszewski, 2017, Edo, Skoglund, Gao, Persson and Jansson, 2016, Wild, Deutmeyer, Bradley, et al., 2016)
	Hydrothermal treatment	A reaction of alkali metals with organic chlorides under high temperatures, producing inorganic Cl, which can be water washed and dewatered	Removes only *organic* Cl and increases the production cost of SRF.	(Prawisudha, Namioka and Yoshikawa, 2012)

	Measures	**Description**	**Demerits**	**Reference**
		reducing [Cl] and retaining the calorific value of the fuel.		
	Optimization of waste processing unit operations in MBT/MT plants	Efficiency of mechanical unit operations is far from perfect and optimization of material flow, waste sorting, transportation, storage and safe handling of the materials from the source to final treatment is required	So far, optimization of sorting processes requires more energy demanding plant configurations, adversely affecting sustainability. MBT/MT plants need an economic incentive policy.	(Bayard, Araújo Morais, Ducom, et al., 2015, Fei, Wen, Huang and De Clercq, 2018, Hinkel, Blume, Hinchliffe, Mutz and Hengevoss, 2019, Velis, Longhurst, Drew, Smith and Pollard, 2010)
SRF thermal treatment (co-combustion in cement kilns)	Operating improvements	Minimization of gas temperature fluctuations, enhancement of gas mixing with gas recirculation, sufficient residence time and combustion temperature	Hard to achieve when highly inherent heterogeneous materials are used as co-fuels.	(Lee, Themelis and Castaldi, 2007, Rademakers, Hesseling and Van de Wetering, 2002, Schorcht, Kourti, Scalet, Roudier and Sancho, 2013)
	Co-combustion at a conservative substitution rate	Depending on the fuel quality and compatibility of SRF properties with the thermal recovery process (50% is a realistic value for cement kilns)	Less cost-effective solution than the optimum rate.	(Gendebien, Leavens, Blackmore, et al., 2003, Velis, Longhurst, Drew, Smith and Pollard, 2010)
	Installation of Cl-bypass system	Details are given in sub-section 7.1.4	High investment cost, which indicatively can reach 5 million US$.	(Enders and Haeseli, 2011, Lechtenberg and Diller, 2012)

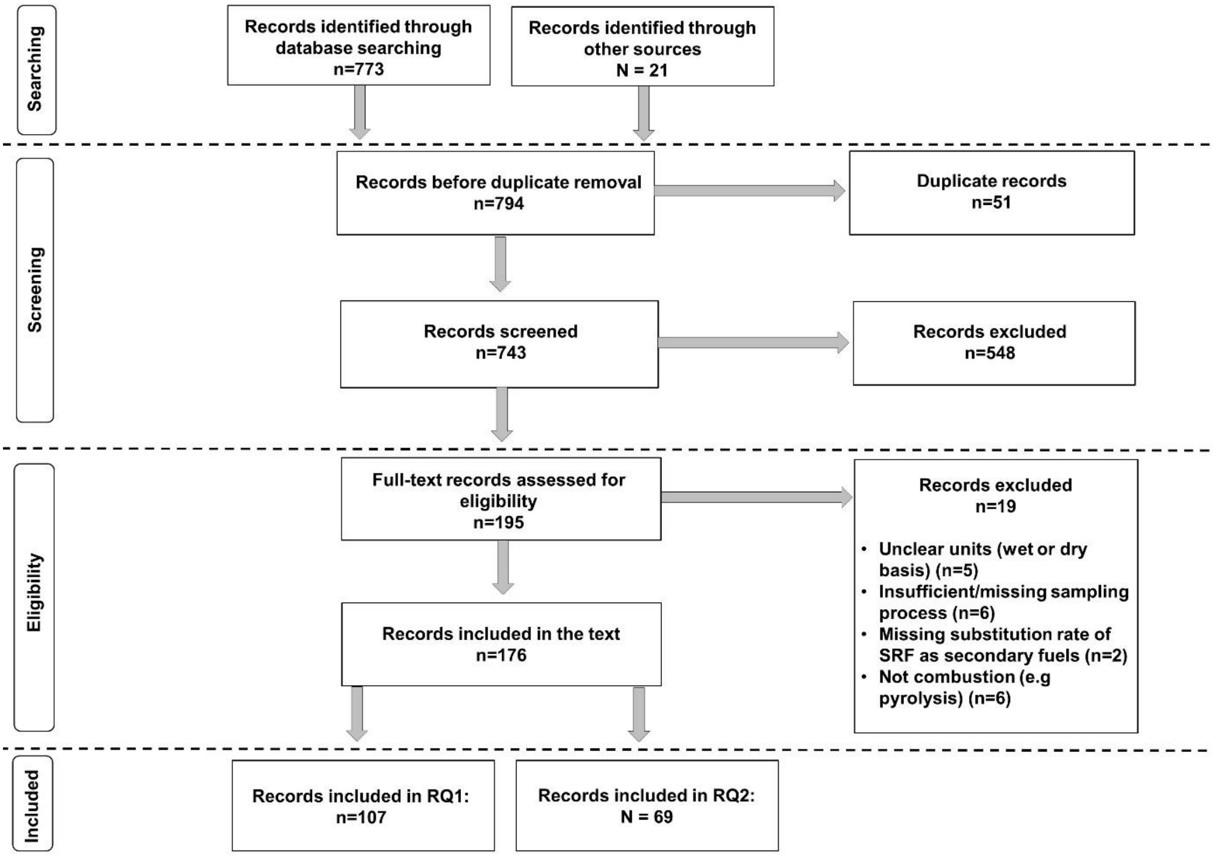

Fig. 1. Flow of information through the different phases of systematic review (PRISMA flow diagram) for Cl in SRF co-combusted in cement kiln systems.

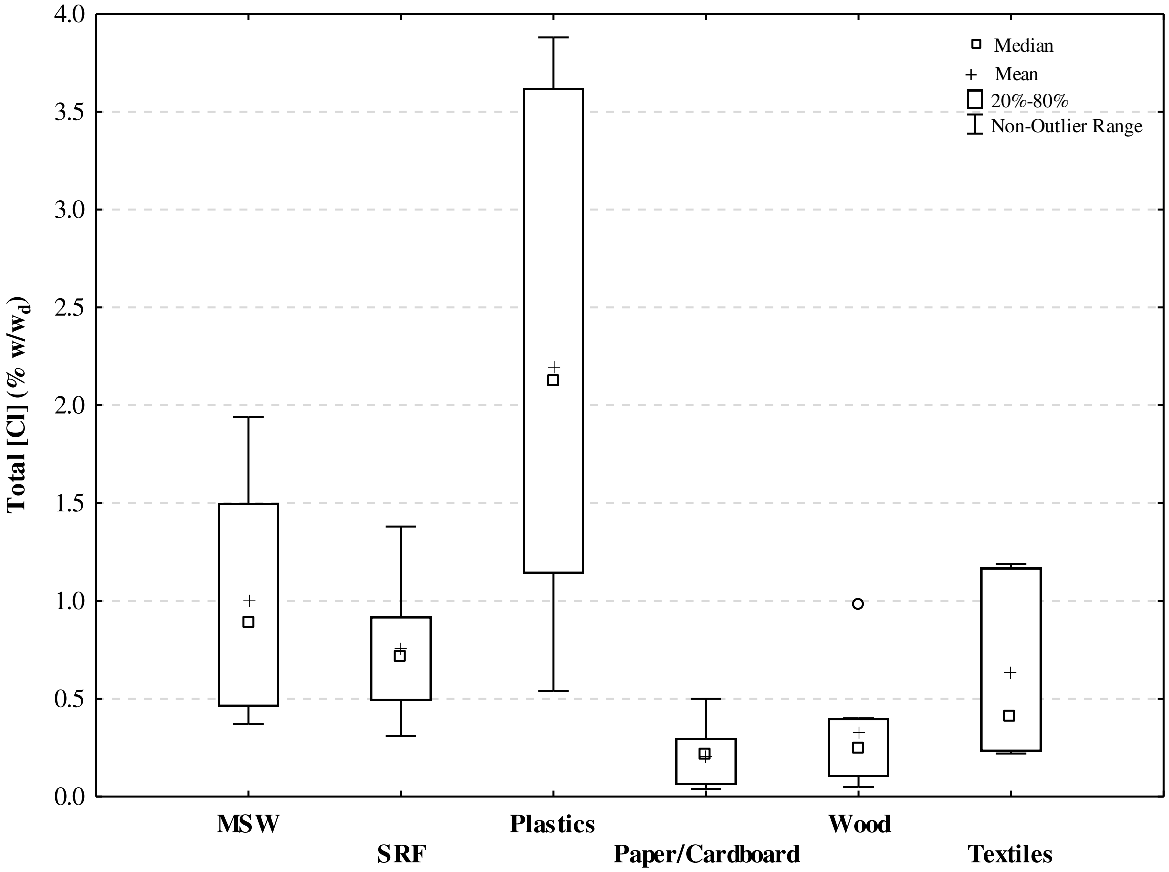

Fig. 2. Boxplots of Total Cl concentration in MSW, SRF and waste sub-fractions (component categories) data. Note: component 'rubber-leather' was excluded due to data limitations and inconsistencies.

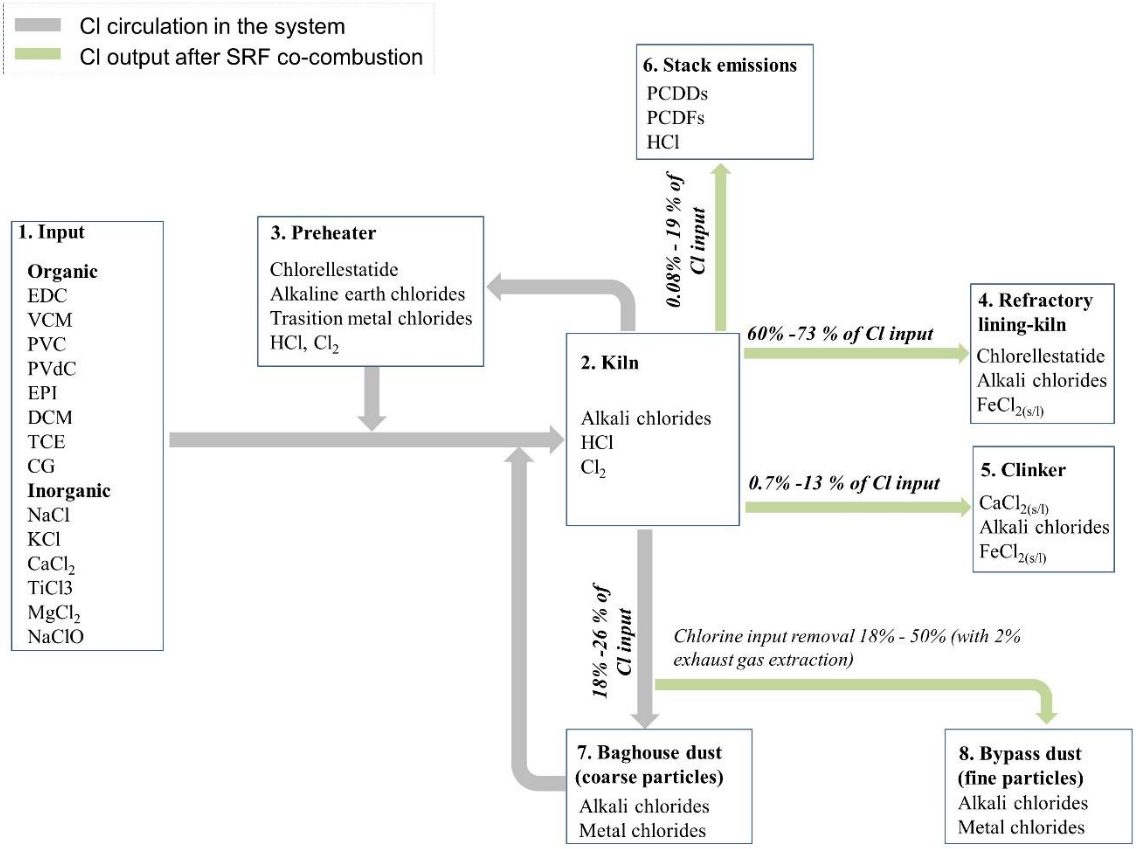

Fig. 3. Most prevalent chloride compounds of SRF and their fate during co-combustion in a kiln-preheater system: Input chlorinated compounds present in SRF **(1)** are volatilized during combustion in the kiln **(2)** and are condensed in the preheater **(2)** forming a Cl circulation, by which Cl can remain in the kiln ('stock') **(4)**; leave the system through the clinker **(5)** or as unburned chlorinated emissions in the stack **(6);** and concentrated in CKD (coarse particles are returned to the kiln **(7)** whereas fines can be removed from the system if bypass system installed **(8)**.

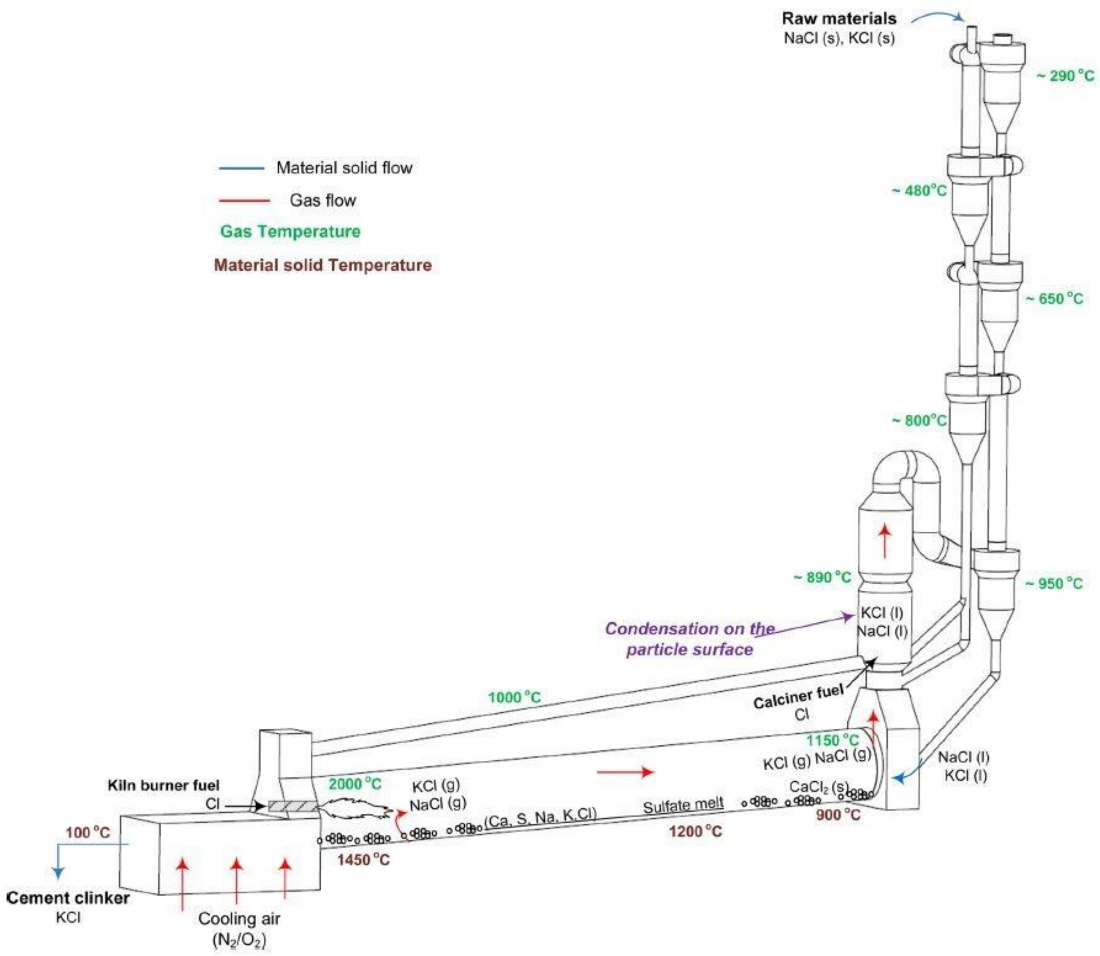

Fig. 4. Chlorine circulation in a modern cement kiln: Cl volatilization occurs in the burning zone at a temperature range of 900°C to1200°C and Cl condensation occurs in the lower cyclones of the preheater forming mixtures with low melting points, which return to the high temperature zone, creating a continuous volatilization-condensation reaction. Adopted from: Cortada Mut, Nørskov, Frandsen, Glarborg and Dam-Johansen (2015).

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