Title Page

Title: Life cycle assessment of pyrolysis, gasification and incineration waste-to-energy technologies: theoretical analysis and case study of commercial plants

Authors: Jun Dong ^{1, 2, *}, Yuanjun Tang ^{1, 2}, Ange Nzihou ¹, Yong Chi ², Elsa Weiss-Hortala ¹, Mingjiang Ni ²

Affiliation: ¹ Centre RAPSODEE, Ecole des Mines Albi, Campus Jarlard, 81013 Albi Cedex, France

² State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou, China

Corresponding author: Jun Dong (Tel.: +33-0563493241; fax: +33-0563493243; E-mail address: zd_dongjun@zju.edu.cn)

Abstract: Municipal solid waste (MSW) pyrolysis and gasification are in development, stimulated by a more sustainable waste-to-energy (WtE) option. Since comprehensive comparisons of the existing WtE technologies are fairly rare, this study aims to conduct a life cycle assessment (LCA) using two sets of data: theoretical analysis, and case studies of large-scale commercial plants. Seven systems involving thermal conversion (pyrolysis, gasification, incineration) and energy utilization (steam cycle, gas turbine/combined cycle, internal combustion engine) are modelled. Theoretical analysis results show that pyrolysis and gasification, in particular coupled with a gas turbine/combined cycle, have the potential to lessen environmental loadings. The benefits derive from an improved energy efficiency leading to less fossil-based energy consumption, and reduced process emissions by syngas combustion. Comparison among the four operating plants (incineration, pyrolysis, gasification, gasification-melting) confirms a preferable performance of the gasification plant attributed to syngas cleaning. The modern incineration is superior over pyrolysis and gasification-melting at present, due to the effectiveness of modern flue gas cleaning, use of combined heat and power (CHP) cycle, and ash recycling. Sensitivity analysis highlights a crucial role of the plant efficiency and pyrolysis char land utilization. The study indicates that the heterogeneity of MSW and syngas purification technologies are the most relevant impediments for the current pyrolysis/gasification-based WtE. Potential development should incorporate into all process aspects to boost the energy efficiency, improve incoming waste quality, and achieve efficient residues management.

Keywords: Waste-to-energy technology; Environmental sustainability; Life cycle assessment; Non-toxic and toxic impacts; Large-scale commercial plants; Improvement and impediments

Main text

1. Introduction

In the transition towards more sustainable development, treatment technologies for municipal solid waste (MSW) have made considerable progress (Zhao et al., 2009). The last decades witnessed a gradually decreased proportion of landfill as required by the European Landfill and Waste Framework Directives (Council of European Communities, 1999, 2008). In contrast, waste-to-energy (WtE) is gaining increasing interest. Until recently, incineration is the most widespread WtE technology with more than 1400 incineration plants in operation around the world (Leckner, 2015). However, even the last generation of MSW incinerators is limited by a low electricity efficiency up to about 22-25% (Panepinto et al., 2015), due to the limitation in the maximum steam temperature of the boiler, normally less than 450 °C to prevent corrosion by gaseous HCl (Belgiorno et al., 2003). Although modern and well-operated incinerators can fulfil the requirements of an environmentally sound technology, potential risk of PCDD/Fs still present as a debate for the public. As a consequence, technological development towards more environmental-friendly and energy-efficient alternative WtE options are still required.

In recent years, there is considerable interest in new WtE technologies particularly pyrolysis and gasification, which attain the possibility to obtain a syngas suitable for different applications (Funari et al., 2016; Khoo, 2009). About energetic use in WtE plants, there is a general perception that pyrolysis and gasification could achieve a higher efficiency by supplying the syngas with a more efficient energy conversion

device such as a gas turbine/combined cycle (gas turbine/CC) or an internal combustion engine (Arena, 2012). Even if in a steam cycle plant, the limitation of efficiency could be overcome by adding gas pre-treatment before it goes into the burner, to allow the removal of HCl and an improvement in steam temperature of 520-540 °C (Belgiorno et al., 2003). Besides, pyrolysis and gasification have the potential to diminish PCDD/Fs (Noma et al., 2012), thus reducing the total generation of pollutants if the downstream syngas oxidization is processed efficiently. However, using of the newly developed WtE options does not automatically guarantee the total sustainability of the whole multi-stage thermal conversion and energy utilization chain (Ning et al., 2013; Wang et al., 2015). The “raw” syngas, which contains a variety of contaminants such as H₂S, tar, NH₃ and particulate matter (PM), needs to be purified to meet the stringent requirement of entering an engine (Wood et al., 2013). The configuration of different energy cycles downstream may also influence the overall environmental effects: the consumptions and losses of gasification and syngas clean-up may cause the overall energy efficiency be close or lower to incineration. It is not a simple procedure to select an optimal WtE technology. A comprehensive assessment of different WtE process configurations is necessary to understand if pyrolysis/gasification-based WtE may become potential alternative or improvement for the current incineration.

Guided by ISO standards (ISO, 1997), life cycle assessment (LCA) is benefited from the quantification of the entire life cycle impacts. This can help identify the most critical process for environmental burdens (Millward-Hopkins et al., 2018), and

provide a benchmark for new technologies. LCA has provided reliable evaluation of MSW treatment technologies (Kaplan et al., 2009; Lundie and Peters, 2005; Morselli et al., 2008; Wäger and Hischer, 2015). However, LCA of WtE technologies is rarely performed other than incineration. This is mainly because the operational practice using pyrolysis and gasification is quite limited despite that a number of applications do exist (Molino et al., 2016; Panepinto et al., 2015), making comparisons very difficult. The existing studies are focused mainly on the thermal conversion process itself, while few of them examine the downstream use of syngas in detail. The environmental performance of WtE options depends on many factors such as emission levels, energy efficiencies, type of end-use applications, and energy source. However, the LCA studies available on pyrolysis and gasification are often based on varying assumptions and insufficient to thoroughly study these issues. This may limit the LCA comparisons between different WtE technologies on a consistent and common basis.

The goal of this work is to provide a detailed life cycle investigation of different WtE technologies. In response to the incompleteness and scarcity of data on pyrolysis and gasification, this study is striving to conduct both theoretical analysis of the possible configuration of WtE technologies and real case studies of several commercial plants. In the first part, a general and extensive theoretical analysis of seven multi-stage WtE systems involving thermal conversion (pyrolysis, gasification, incineration) and energy utilization (steam cycle, gas turbine/CC, internal combustion engine) is modelled, using the most typical and well-accepted reported data. In the second part, four large-scale commercial operation WtE plants (pyrolysis, gasification,

gasification-melting, modern incineration) are compared. Besides, a sensitivity analysis is carried out to identify key parameters responsible for the environmental impacts. This study aims at understanding how the current WtE could get a benefit towards a more environmentally sustainable technology. Potential improvements and impediments to the further development of pyrolysis and gasification-based WtE technologies are also discussed and suggested.

2. Methodology

2.1. System definition

The system boundaries (**Fig. 1**) of the study attain at the moment when MSW enters the WtE plant. Four basic processes are included: (1) MSW pre-treatment, (2) thermal conversion, (3) utilization of acquired products, and (4) ash and air pollution control (APC) residues management. MSW can either be thermally converted by adding sufficient amount of air (incineration), where the MSW is fully oxidized into process heat; or by supplying an air deficiency, where the waste is pyrolyzed (in the absence of air) or gasified (in a partial oxidant amount lower than stoichiometric combustion). The latter case produces intermediated products including syngas, tar and char, which can recover energy in several pathways (Molino et al., 2016): to be combusted in a boiler and connected with a steam turbine; or, after a purification step, to be used in a gas turbine/CC or an internal combustion engine. Thus a total of seven scenarios are formed. S1 is defined as MSW direct incineration to represent the current WtE. S2, S3 and S4 represents pyrolysis coupled with steam turbine, gas turbine/CC and internal

combustion engine, respectively; gasification combined with those energy devices are defined as S5, S6 and S7. MSW pre-treatment mainly refers to drying and shredding with the aim of size reduction and homogenization. While incineration plants could process MSW directly (Evangelisti et al., 2015), pre-treatment is basically needed prior to pyrolysis/gasification (McKendry, 2002). Detailed flowchart of each system is illustrated in the Supplementary Material (**Fig. S1**).

The functional unit is set at one ton of MSW as received at the plant. Upstream production of fuels and materials including diesel, electricity, lime, etc. is considered as the ‘cradle to grave’ type of calculation. The benefits from useful co-products, such as electricity and heat, are allocated by system expansion. The recovered electricity is assumed to substitute that provided by the “energy mix” of a specific region, here the European average (42.7% fossil fuels, 26.5% nuclear, 30.0% renewable energies, 0.7% waste and 0.1% other in 2015) is selected (Eurostat). The produced heat displaces an equal amount of heat generated by “heat mix”, of which the heat production data based on European average is again used (69.3% fossil fuels, 0.2% nuclear, 22.9% renewable energies, 4.8% waste and 2.7% other in 2015) (Eurostat). The database Gabi 7.0 provides the remaining, mainly indirect burdens, of the background system.

2.2. MSW characteristics

The MSW typically treated in the WtE plant is the residual from the source-separated collection of dry recyclables and wet organic fractions. The waste

characteristics in different countries have a high variability depending on the culture, climate and socioeconomic (Vergara and Tchobanoglous, 2012). Therefore, a typical MSW, reflects the average waste composition in Europe (Arena et al., 2015), is selected as the basis for comparison (**Table 1**).

2.3. Data source for theoretical analysis

The data utilized for theoretical analysis are mainly derived from industrial practice, peer-reviewed literature, standards, and recent research reports. The data are regionalized in the sense that they refer to the situation of Europe. For each of the WtE system, the modelling of material consumptions, emissions and energy recovery is analyzed on basis of mass and energy balance, the detailed calculations are available in the Supplementary Material (**Section SM-8**). Each unit process and the main data source are presented as following.

2.3.1. MSW pre-treatment

A pre-treatment step is assumed to be conducted before pyrolysis and gasification process. In order to facilitate homogenization, the incoming MSW is shredded to an average size of a few hundred millimeters (e.g. a size of around 100 mm in practice). The estimated energy use for mechanical treatment is set at 100 kWh of electricity and 25 kWh of natural gas per ton of MSW (Kourkoumpas et al., 2015). The waste then undergoes drying to a final moisture content of around 10%. The heat required by the dryer is internally supplied with a thermal efficiency of 90% (Roberts et al.,

2009). For systems using gas turbine/CC and internal combustion engine (S3, S4, S6 and S7), the heat derives from the syngas purification unit which recovers the sensible heat of the hot syngas during cooling. For S2 and S5, the heat is supplied by the hot flue gas.

2.3.2. Thermal conversion (pyrolysis, gasification and direct incineration)

For pyrolysis, the proportion of each product (syngas, tar and char) is strongly dependent on the reaction temperature, residence time and heating rate (Van de Velden et al., 2010). For waste processing, a running temperature of 500-550 °C is widely used in industrial plants (Chen et al., 2015). This pyrolysis technology, represented by the RWE-ConTherm[®] process (Hauk et al., 2004), is considered in this analysis, since it is the most typical pyrolysis process presently available in the European market. The pyrolysis reactor is a rotary kiln type, with a residence time of approximately 1 hour. About 85% of the energy will be converted into the hot gas (i.e., hot gas efficiency), with cold gas efficiency attaining around 50%. The cold gas efficiency can be defined as the ratio of the energy content of the cold syngas to that of the feedstock. The balance is char, and its mass proportion is around 30%. The data are based on average reported values of the industrial plants (DGEngineering - The rotary kiln engineers, July 2009a, b). We assume the reliability is high because they can be cross-checked extensively.

Gasification owns the sole objective to produce syngas, although the generation of tar is inevitable along with the gas. In comparison to pyrolysis, gasification occurs at a

generally higher temperature: 550-900 °C in air gasification and 1000-1600 °C if using pure oxygen, oxygen-enrich gas or steam (Arena, 2012). Based on several operation data from the existing plants, the cold gas efficiency is in a range of 50-80% (Arena, 2012). Here a cold gas efficiency of 70% is used as a conservative estimate (Panepinto et al., 2015; Yassin et al., 2009). A hot gas efficiency of 90% is assumed in the case syngas is directly used in a boiler without any pre-cooling.

MSW direct incineration is well-proven and has greater operational reliability than pyrolysis and gasification. The assumed incineration is based on a moving grate. The waste is directly combusted to heat up water in the boiler to generate steam. A heat loss is also inevitable, for example the discharge of the ash and flue gas will cause a high loss of the sensible heat. However, we do not tend to assume this efficiency, since it will be reflected in the overall plant efficiency.

While the incineration process is exothermic, gasification can also achieve heat self-sustaining around an equivalence ratio of 0.3-0.4 (Zhang et al., 2011), i.e. no any external thermal assistance is needed, the same under which in the real plants (Arena and Di Gregorio, 2014). Nevertheless, pyrolysis requires an additional thermal energy to maintain the reaction. The input energy is around 9% of the MSW energy according to the research of Baggio et al. (Baggio et al., 2008). The heat is assumed to be supplied by the hot flue gas as it is commonly preferred in the plants.

2.3.3. Energy utilization cycles

Each WtE plant in this study is assumed to be an integrated facility, in which the

final energy utilization is operated on-site. The electrical efficiency of the incineration plant is set at 22.5% (Arena, 2012; Morris and Waldheim, 1998), which represents an average of the modern dedicated waste combustion systems. For pyrolysis and gasification systems, steam cycle is the simplest option because the hot syngas could undergo combustion in the gas boiler without purification. A higher efficiency can be achieved (set at 27.8% in this study), since the homogenous and gas-phase combustion is more controllable and effective (Consonni and Viganò, 2012). The syngas can also be burned in a gas turbine/CC or an internal combustion engine. Potentially, the electrical efficiencies would be higher (set at 35.5% and 25.0% for gas turbine/CC and engine, respectively (Belgiorno et al., 2003; Morris and Waldheim, 1998)). However, the syngas needs to be cooled and purified to meet the stringent inlet gas quality requirement. To ensure the transparency of the data, the values of plant efficiencies are determined by extensively searching and comparing with similar set-up in the literature and reports (see details in **Table S2**). Additionally, a range of variations of each plant efficiency will be discussed in the sensitivity analysis. For all the systems analyzed, 20% of the generated electricity is assumed to be self-consumed in the plant, with the remaining 80% sent to the power grid.

For systems using gas turbine/CC and internal combustion engine (S3, S4, S6 and S7), cleaning the syngas allows the chemical energy to be conserved. The sensible heat is recovered assuming an efficiency of 75% (Yi et al., 2013). As stated earlier, the heat is used for MSW pre-treatment; the excessive amount is transferred to the needs of the end user. The formed pyrolysis char can either be combusted at the facility to

generate more energy or be used as a product (biochar). The former application is considered as the baseline, while the latter case will be discussed in the sensitivity analysis. The pyrolysis char is assumed to be sent into the boiler and combusted together with the gas in the S2 system, which is in accordance with the real operation in reference plants. If a gas turbine/CC or internal combustion engine is used, the char is assumed to be combusted in a separated boiler for heat production at a thermal efficiency of 75%, which is a typical value for industrial heating boilers in operation (Roberts et al., 2009).

2.3.4. Emissions at the stack

In attempt to better perform a transparent evaluation, the emission factors used in this theoretical analysis are estimated using the European pollution control standards, i.e., the exhaust flue gas from each WtE system is assumed to meet the requirements of specified emission standards (Directive 2007/76/EC (The Commission of the European Communities, 2007) and Directive 2010/75/EU with some adaptations (Directive, 2010)). The real emission data from industrial plants will be analyzed in the second part (case studies). **Table 2** summarizes the related emission factors. These data have been used in conjunction with estimates of flue gas volumes per functional unit of MSW produced to derive the final mass release rates. Details on the standards, adaptations and calculations can be found in the Supplementary Material (**Section SM-5**).

2.3.5. Ash and air pollution control residues management

The amount of solid residues produced by incineration and pyrolysis/gasification plants are assumed to be 180 kg/t-MSW and 120 kg/t-MSW, respectively, as reported by UK's waste report (DEFRA UK, 2004). The solid residues may be recycled as road construction materials or concrete aggregate (Sakai and Hiraoka, 2000). However, only landfill is considered in the theoretical analysis and the potential benefit will be included in the case studies of the commercial plants. The APC residues, including mainly fly ashes and exhausted sorbents, are assumed to be stabilized before final disposal in landfill. Emissions, mainly heavy metals to the soil, are estimated according to the UK's waste report (DEFRA UK, 2004).

2.4. Data source for commercial operation WtE plants

Four large-scale commercial operation WtE plants (pyrolysis, gasification, gasification-melting, modern incineration) are modeled as case studies. The selected plants could represent the most typical modern state-of-the-art plants, therefore reflecting the actual environmental sustainability of different WtE technologies. The selected plants are all in connection with a steam turbine cycle, i.e., in a similar configuration of the S1, S2 or S5 system. **Table 2** and **Table S5** summarizes the related emission factors and information of these plants, respectively; with a brief introduction of each plant presented as following.

●**Incineration plant (C1)**: Silla 2 incineration plant, located in Milan, Italy, is studied as a typical case of the modern incineration. The plant is equipped with 3

moving grate combustion lines, having a treatment capacity of 450,000 t/a. MSW is incinerated at 850 °C to produce electricity and district heating at an efficiency of 24% (net) and 6%, respectively (Turconi et al., 2011). The flue gas cleaning includes electrostatic precipitator, acid gas neutralization (NaHCO₃ injection), fabric filter and a SCR unit for NO_x abatement (Amsa, April, 2008). After combustion, metals are sorted from the bottom ash and recycled. 88% of the bottom ash is utilized in road construction, while the remaining fraction is landfilled and the APC residues are safely disposed.

●**Pyrolysis plant (C2):** The selected plant, located in Hamm, Germany, has a capacity of 100,000 t/a, although it is no longer in operation after the chimney collapse in 2009. The pyrolysis process belongs to the RWE-ConTherm[®] technology (DGEEngineering - The rotary kiln engineers, July 2009a). After shredded to 200 mm, the MSW is decomposed in the absence of air in a rotary kiln at 500 °C with a residence time of 1 hour, using natural gas as the heating source. The products, hot syngas and char, are incinerated in the boiler of a coal-fired plant for electricity production. The residues are considered to be landfilled and the metals are recycled. The plant electricity efficiency (gross) is around 22% (Stein and Tobiasen, March 2004).

●**Gasification plant (C3):** The selected plant, Lahti II, located in Finland, has started its commercial operation in 2012 with an annually capacity of 250,000 tons (Lahti Energia). The feedstock is solid recovered fuels (SRF), i.e., high calorific waste unsuitable for recycling. The gasifier is a circulating fluidized bed operated at

850-900 °C. The syngas generated undergoes cooling at 400 °C to remove heavy metals and PM. The cleaned syngas enables a more efficient heat recovery boiler at 121 bar and superheated steam at 540 °C. The plant attains final 27% of electricity efficiency (net) and 61% of heat efficiency (Savelainen and Isaksson, 2015). The flue gas cleaning system consists of a bag house filter with additive injections (NaHCO₃ and activated carbon) and a SCR for NO_x reduction. From the plant outlet, the bottom ash is removed to landfill disposal and the APC residues are safely disposed.

●**Gasification-melting plant (C4)**: The reason to select this technology is its possibility to recover materials effectively (Tanigaki et al., 2012). The selected plant, having a total throughput of 80 MW, is located in Japan and is one of the largest gasification-melting facilities in the world. The MSW is charged into a shaft-furnace type gasifier from the top with coke and limestone, and the ash is melted at the bottom by O₂-rich air at 1000-1800 °C. No pre-treatment of the incoming waste is required. The syngas is transferred to be combusted to generate steam at 400 °C and 3.92 MPa. The electricity efficiency (gross) attains at 23% (Tanigaki et al., 2012). The flue gas cleaning applies a quencher, a baghouse with Ca(OH)₂ injection for desulfurization, a re-heater and a SCR for NO_x reduction. The molten materials from the gasifier are magnetically separated into slag and metals, which can be completely recycled; while the APC residues are further treated.

2.5. Life cycle inventory

By combining all unit processes input-output data, a detailed LCI table is compiled

(see **Table S6** and **Table S7**). Biogenic CO₂ is assumed to be carbon neutral to global climate change. For the specific MSW in this study, the fraction of biogenic carbon contributes 64% of the received MSW. Emissions to the water are not included, since modern WtE systems are commonly designed with wastewater treatment and reused equipment to meet a ‘zero discharge’ target (Chen and Christensen, 2010).

2.6. Life cycle impact assessment

The well-accepted Danish EDIP methodology is used to aggregate the LCI data (Hauschild and Potting, 2005; Wenzel et al., 1997). Seven impact categories are considered: global warming (GW), acidification (AC), terrestrial eutrophication (TE), photochemical ozone formation to human health (POFh), human toxicity via air (HTa) and solid (HTs), and ecotoxicity via solid (ETs). Results based on normalized values are used to reflect the relative magnitude of different impacts into person equivalence. A summary of the normalization references is available in **Table S8**.

3. Results

3.1. Theoretical analysis results

Fig. 2 reports the overall environmental performance of different systems. Compared to direct incineration (S1), pyrolysis and gasification are effective to lessen the environmental impacts of TE, POF, HTa and ETs, yet increase the burdens of GW and HTs. For a direct comparison of different WtE processes, gasification systems (S5-S7) lead to a lower impact than pyrolysis systems (S2-S4). For systems using

different energy cycles, gas turbine/CC (S3, S6) has surpassed steam turbine (S2, S5) and internal combustion engine (S4, S7) and becomes the most preferred energy utilization approach.

To give a clear and transparent explanation of the aforementioned results, the overall impact is divided into four stage-wise contributors: energy input, direct emissions, ash management and energy recovery. As shown in **Fig. 3**, the environmental savings for non-toxic impacts are primarily brought by energy recovery, which compensates a significant amount of emissions generated by fossil fuel-based energy production. In particular, negative values appear for several systems regarding AC, TE and POFh, indicating that the environmental benefit has balanced the loading and a net environmental saving is achieved. The highest recovered energy has been found for systems equipped with gas turbine/CC (S3, S6). This reveals the advantage brought by a more efficient energy device that is able to counterbalance an increasing amount of emissions. Besides, pyrolysis equipped with combustion engine (S4) also exhibits significant avoided impacts due to the additional savings from process heat (mainly from tar and char combustion), which highlights the importance of heat recovery in improving the total recovered energy.

Direct emissions also have a large influence to the total impacts. Different systems show negligible difference of GW, because CO₂ emission is decisively contributed to GW and it mainly derives from the fossil-origin carbonaceous compounds contained in MSW. However, there is a dramatic difference in direct emissions among all the systems, if consulting the impacts of AC, TE and POFh. Compared with incineration

(S1), 21-34% and 28-83% decrease in those indicators are achieved for pyrolysis and gasification systems, respectively. The principal contributors for AC, TE and POFh are acid gases including NO_x, SO₂, HCl and HF. The reduced emissions by pyrolysis and gasification can in fact be ascribed to, on one hand, a lower amount of flue gas as a consequence of the lower excess air required for syngas combustion; on the other hand, the limited NO_x generation as a result of the homogeneous gas-gas reaction (Consonni and Viganò, 2012). It shows also a further reduction of emissions from gasification systems using gas turbine/CC and internal combustion engine (S6, S7), because purifying the syngas allows the removal of a part of acid gases; and, the syngas volume is much smaller to limit the total flue gas. Conversely the direct emissions from pyrolysis systems (S3, S4) tend to increase due to char and tar combustion.

All systems contribute positive impacts to toxic categories including HTa, HTs and ETs. **Fig. 2** reveals that HTs and HTa are the highest burden categories, being 1-2 orders of magnitude more significant than non-toxic impacts. Direct emissions and ash management are the main contributors. The avoided emissions are insignificant, which is opposite to that of non-toxic impacts. The toxic impacts are decisively due to heavy metals, PCDD/Fs and PM emissions for their relatively high equivalent factors. Ash management takes a crucial effect to HTs and ETs, since heavy metals contained in the ash is liable to be transferred into the soil after landfill, or released during the solidification/stabilization process of the APC residues.

Consequently, it could be concluded from the theoretical analysis that compared

with incineration, both pyrolysis and gasification own the potential to have a better environmental performance due to two-folds benefits: the reduced process emissions as well as a substantial increase in the amount of energy recovered. However, the important input energy demand, for example waste pre-treatment, syngas cleaning and endothermic pyrolysis reaction, may on the other hand become additional burdens especially regarding GW. This is also one reason for an inferior performance from pyrolysis systems in comparison to gasification. Overall, gasification equipped with gas turbine (S6) is observed to be the most environmentally preferable system.

3.2. Case studies for commercial operation WtE plants

Fig. 4 summarizes the environmental impacts from four large-scale commercial operation WtE plants, where all impacts experience a significant drop compared with the theoretical analysis. The benefit is mainly due to the reduction in the process direct emissions, revealing that plants based on all the technologies in connection with a steam boiler can comfortably meet the required emission limits. The environmental sustainability of each plant in descending order is: gasification > incineration > (pyrolysis, gasification-melting); while it is difficult to figure out the relative superiority between pyrolysis and gasification-melting. It reveals that the modern incineration could fulfil an environmentally sound technology, i.e., better than pyrolysis and gasification-melting plants at present. The emission factors reported in **Table 2** indicate that the actual emissions from the reference incineration and pyrolysis/gasification plants are quite similar due to the technological performance of

the modern flue gas cleaning devices (fabric filters, desulfurization, NO_x abatement, activated carbon injection, etc.). The improved performance of incineration could also be attributed to the use of the more efficient combined heat and power (CHP) cycle, which has achieved an additional 6% of heat production. On the other hand, gasification reaches the best performance among the four plants. This fact again verifies the positive role of syngas cleaning, which allows the gas clean enough to employ higher steam data (540 °C, 121 bar compared with 400 °C, 40 bar in conventional waste boiler) for an increased electricity efficiency (27% net compared with 24% in incineration). Additionally, this gasification plant shows further advantage by an abundance of heat production (61%), significantly larger than in the incineration plant (6%). Those together have resulted in a significant environmental saving from the avoided heat and electricity production in the gasification plant.

Contrarily, pyrolysis and gasification-melting plants show an inferior performance. The increased environmental burdens are either due to a high amount of auxiliary fuel used, or a low amount of net energy recovered. Particularly, gasification-melting plant shows an important internal parasitic energy demand reaching 24% of the total energy production, mainly due to the use of O₂-rich air for ash melting.

An obvious reduction in HTs and ETs is achieved in the incineration and gasification-melting plants. The offset impacts are mainly attributed to the recycling of the bottom ash, slag and metals owing to two-aspects benefits: the reduced amount of ash to be treated, which is the main cause of solid heavy metals leaching; and, the avoided manufacture of road construction materials and metals from their virgin

materials.

3.3. Sensitivity analysis

A sensitivity analysis has been carried out to identify key process parameters as well as to seek for potential improvements. The evaluation is based on the data from the theoretical analysis considering two variations: changing of the plant efficiency, and alternative utilization of the pyrolysis char as soil amendment.

A $\pm 10\%$ variation of the plant efficiency for each system is conducted. Results in **Table 3** show an up to $\pm 665\%$ variation in the environmental impacts, of which non-toxic impacts appear of remarkable relevance. The variation is primarily related to the amount of energy recovered as it could replace the associated emissions from the burning of fossil fuels. The results confirm a crucial role of the energy recovery efficiency in determining the total sustainability of a WtE plant.

For pyrolysis systems (S2-S4), the sensitivity analysis considers also the case where the char is used as soil amendment. In such case, the pyrolysis char is considered to have two additional merits (Harder and Forton, 2007; Roberts et al., 2009): substitution of fertilizer (N, P and K) and carbon sequestration. Key assumptions and calculations are presented in **Table S10**. **Fig. 5** indicates that this assumption has exhibited an obvious reduction on the majority of impacts except for HTs and ETs. The benefit is dominantly attributed to the reduced airborne emissions from char combustion, together with a small portion of avoided emissions from fertilizer substitution and carbon sequestration. However, a non-negligible increase of

the HTs and ETs loadings are observed due to the increased heavy metals to soil, which should be controlled effectively apart from the associated potential benefits of land application.

4. Discussions

Pyrolysis and gasification have been applied to waste treatment since 1970s, however their commercial application does not achieve widespread so far (Panepinto et al., 2015). One of the main impediments is the heterogeneity of MSW, i.e., inconstant on size and highly variable on composition, which could not easily run stable. Despite this challenge, after years of practical experience, the main technical difficulties seem to be solved and innovative plants started to be operated (Panepinto et al., 2015).

The theoretical analysis of this study shows that using pyrolysis/gasification to supply a gas turbine/CC may achieve higher energy efficiencies and lower emissions than the current incineration. However, its application has not yet overcome many obstacles. For example, the state-of-the-art syngas purification technologies do not achieve the required quality standards. Also running gas turbines require complex maintenance. These reasons have in fact caused a very limited application of the gas turbine/CC in pyrolysis/gasification-based WtE plants (Panepinto et al., 2015); while the most common configuration today is to burn the syngas in a steam boiler, namely, “two-step oxidation” (Consonni and Viganò, 2012).

In recent years, development of the pyrolysis/gasification-based WtE technologies

has become a focus of attention, stimulated by the search for more efficient energy recovery and environmentally sustainable waste management. However, case studies results based on the current large-scale commercial plants reveal that the modern incineration could fulfil an environmentally sound technology, which performs better than the selected pyrolysis and gasification-melting plants. To be commercially successful, the pyrolysis/gasification-based WtE must develop the whole process chain (pre-treatment, thermal conversion, products utilization, residues management).

Those potential areas of development could include:

1. Boost the plant efficiency. The superior performance of the Lahti gasification plant attains at its effective syngas cleaning, which facilitates increasing the steam parameters while avoiding the corrosion problem. It could serve as a demonstration for designing the next generation of WtE configuration. The overall energy efficiency could also be increased by the utilization of the CHP system, or syngas co-incineration in a higher efficiency power station.

2. Use of selected waste streams. Pyrolysis and gasification plants tend to require very careful feedstock pre-treatment. To be more effective, solutions could be the use of SRF, refuse derived fuel (RDF), or residuals from mechanical biological treatment (MBT) systems, which are more homogenous than the raw MSW.

3. Efficient residues management. Recycling materials from WtE solid residues, particularly metals and bottom ash, may result in two main benefits: a decrease in waste landfill; and, a reduction in the consumption of virgin raw materials. Pyrolysis plant could also consider the use of char in land application. The specific properties of

bottom ash/char, in particular the leaching behaviour, should be carefully considered to ensure that the residues would not cause adverse environmental impacts.

Long-term potential areas of development could also attain at (Engineers, 2004):

1. Syngas purification and use in higher energy efficiency equipment such as a dedicated gas turbine/CC.

2. Further processing of syngas to be used as chemical feedstock, liquid fuels, etc.

Acknowledgments

This project is supported by the French Centre National de la Recherche Scientifique (CNRS), the Labex SOLSTICE (Agence Nationale de la Recherche, ANR), and the Chinese Program of Introducing Talents of Discipline to University (B08026).

Appendix A. Supplementary data

Supplementary data to this article (detailed process description, data acquisition and calculation, inventory analysis, and the sensitivity analysis used in the LCA) are available.

References

- Amsa, April, 2008. Silla 2 waste to energy plant.
- Arena, U., 2012. Process and technological aspects of municipal solid waste gasification. A review. Waste Manage. 32, 625-639.
- Arena, U., Ardolino, F., Di Gregorio, F., 2015. A life cycle assessment of

485 environmental performances of two combustion-and gasification-based
 486 waste-to-energy technologies. *Waste Manage.* 41, 60-74.

487 Arena, U., Di Gregorio, F., 2014. Gasification of a solid recovered fuel in a pilot scale
 488 fluidized bed reactor. *Fuel* 117, 528-536.

489 Baggio, P., Baratieri, M., Gasparella, A., Longo, G.A., 2008. Energy and
 490 environmental analysis of an innovative system based on municipal solid waste
 491 (MSW) pyrolysis and combined cycle. *Appl. Therm. Eng.* 28, 136-144.

492 Belgiorno, V., De Feo, G., Della Rocca, C., Napoli, R., 2003. Energy from
 493 gasification of solid wastes. *Waste Manage.* 23, 1-15.

494 Chen, D., Christensen, T.H., 2010. Life-cycle assessment (EASEWASTE) of two
 495 municipal solid waste incineration technologies in China. *Waste Manage. Res.*
 496 28(6), 508-519.

497 Chen, D., Yin, L., Wang, H., He, P., 2015. Pyrolysis technologies for municipal solid
 498 waste: a review. *Waste Manage.* 37, 116-136.

499 Consonni, S., Viganò, F., 2012. Waste gasification vs. conventional Waste-To-Energy:
 500 A comparative evaluation of two commercial technologies. *Waste Manage.* 32,
 501 653-666.

502 Council of European Communities, 1999. Directive 1999/31/EC on the landfill of
 503 waste. *Official Journal of the European Union L.*

504 Council of European Communities, 2008. Directive 2008/98/EC of the European
 505 Parliament and of the Council of 19 November 2008 on waste and repealing
 506 certain Directives. *Official Journal of the European Union L.*

507 DEFRA UK, 2004. Review of Environmental and Health Effects of Waste
 508 Management: municipal solid waste and similar wastes. Defra Publications,
 509 London.

510 DGEEngineering - The rotary kiln engineers, July 2009a. Integrated pyrolysis into
 511 power plant, Hamm MW pyrolysis plant.

512 DGEEngineering - The rotary kiln engineers, July 2009b. Waste pyrolysis plant
 513 "Burgau", Burgau MW pyrolysis plant.

514 Directive, E., 2010. Directive 2010/75/EU of the European Parliament and of the
 515 Council of 24 November 2010 on industrial emissions (Integrated Pollution
 516 Prevention and Control). Official Journal of the European Union L 334, 17-119.

517 Engineers, F.C., 2004. The viability of advanced thermal treatment of MSW in the UK.
 518 ETSET report.

519 Eurostat, Gross derived heat production by fuel, TJ, EU-28, 1990-2015
 520 <[http://ec.europa.eu/eurostat/statistics-explained/index.php/File:Gross_derived_h](http://ec.europa.eu/eurostat/statistics-explained/index.php/File:Gross_derived_heat_production_by_fuel,_TJ,_EU-28,_1990-2015_T4.png)
 521 [eat_production_by_fuel,_TJ,_EU-28,_1990-2015_T4.png](http://ec.europa.eu/eurostat/statistics-explained/index.php/File:Gross_derived_heat_production_by_fuel,_TJ,_EU-28,_1990-2015_T4.png)>.

522 Eurostat, Gross electricity production by fuel, GWh, EU-28, 1990-2015
 523 <[http://ec.europa.eu/eurostat/statistics-explained/index.php/File:Gross_electricity](http://ec.europa.eu/eurostat/statistics-explained/index.php/File:Gross_electricity_production_by_fuel,_GWh,_EU-28,_1990-2014-T1.png)
 524 [production_by_fuel,_GWh,_EU-28,_1990-2014-T1.png](http://ec.europa.eu/eurostat/statistics-explained/index.php/File:Gross_electricity_production_by_fuel,_GWh,_EU-28,_1990-2014-T1.png)>.

525 Evangelisti, S., Tagliaferri, C., Clift, R., Lettieri, P., Taylor, R., Chapman, C., 2015.
 526 Life cycle assessment of conventional and two-stage advanced
 527 energy-from-waste technologies for municipal solid waste treatment. J. Clean
 528 Prod. 100, 212-223.

529 Funari, V., Meisel, T., Braga, R., 2016. The potential impact of municipal solid waste
530 incinerators ashes on the anthropogenic osmium budget. *Sci. Total. Environ.* 541,
531 1549-1555.

532 Harder, M.K., Forton, O.T., 2007. A critical review of developments in the pyrolysis
533 of automotive shredder residue. *J. Anal. Appl. Pyrol.* 79, 387-394.

534 Hauk, R., Spindeldreher, O., Usdrowski, N., Stadtmuller, J., Zimmer, F., Marsico, C.,
535 2004. ConTherm-thermal utilisation of waste in a power plant by integrated
536 waste pyrolysis of high-calorific residual derivate fuels. *VGB Powertech* 85,
537 66-71.

538 Hauschild, M., Potting, J., 2005. Spatial differentiation in Life Cycle impact
539 assessment-The EDIP2003 methodology. Institute for Product Development,
540 Technical University of Denmark.

541 ISO, 1997. ISO 14040: Environmental management-Life cycle assessment-Principles
542 and Framework. ISP copyright office, Geneva.

543 Kaplan, P.O., Decarolis, J., Thorneloe, S., 2009. Is it better to burn or bury waste for
544 clean electricity generation? *Environ. Sci. Technol.* 43, 1711-1717.

545 Khoo, H.H., 2009. Life cycle impact assessment of various waste conversion
546 technologies. *Waste Manage.* 29(6), 1892-1900.

547 Kourkoumpas, D.-S., Karellas, S., Kouloumoundras, S., Koufodimos, G., Grammelis,
548 P., Kakaras, E., 2015. Comparison of waste-to-energy processes by means of life
549 cycle analysis principles regarding the global warming potential impact: applied
550 case studies in Greece, France and Germany. *Waste Biomass Valori.* 6, 605-621.

551 Lahti Energia, <http://www.lahtigasification.com/>.

552 Leckner, B., 2015. Process aspects in combustion and gasification Waste-to-Energy
553 (WtE) units. Waste Manage. 37, 13-25.

554 Lundie, S., Peters, G.M., 2005. Life cycle assessment of food waste management
555 options. J. Clean Prod. 13, 275-286.

556 McKendry, P., 2002. Energy production from biomass (part 3): gasification
557 technologies. Bioresource. Technol. 83, 55-63.

558 Millward-Hopkins, J., Busch, J., Purnell, P., Zwirner, O., Velis, C.A., Brown, A.,
559 Hahladakis, J., Iacovidou, E., 2018. Fully integrated modelling for sustainability
560 assessment of resource recovery from waste. Sci. Total. Environ. 612, 613-624.

561 Molino, A., Chianese, S., Musmarra, D., 2016. Biomass gasification technology: The
562 state of the art overview. J. Energ. Chem. 25, 10-25.

563 Morris, M., Waldheim, L., 1998. Energy recovery from solid waste fuels using
564 advanced gasification technology. Waste Manage. 18, 557-564.

565 Morselli, L., De Robertis, C., Luzi, J., Passarini, F., Vassura, I., 2008. Environmental
566 impacts of waste incineration in a regional system (Emilia Romagna, Italy)
567 evaluated from a life cycle perspective. J. Hazard. Mater. 159(2-3), 505-511.

568 Ning, S.-K., Hung, M.-C., Chang, Y.-H., Wan, H.-P., Lee, H.-T., Shih, R.-F., 2013.
569 Benefit assessment of cost, energy, and environment for biomass pyrolysis oil. J.
570 Clean Prod. 59, 141-149.

571 Noma, T., Ide, K., Yoshikawa, J., Kojo, K., Matsui, H., Nakajima, R., Imai, K., 2012.
572 Development of waste gasification and gas reforming system for municipal solid

573 waste (MSW). *J. Mater. Cycles Waste* 14, 153-161.

574 Panepinto, D., Tedesco, V., Brizio, E., Genon, G., 2015. Environmental performances
575 and energy efficiency for MSW gasification treatment. *Waste Biomass Valori.* 6,
576 123-135.

577 Roberts, K.G., Gloy, B.A., Joseph, S., Scott, N.R., Lehmann, J., 2009. Life cycle
578 assessment of biochar systems: estimating the energetic, economic, and climate
579 change potential. *Environ. Sci. Technol.* 44, 827-833.

580 Sakai, S.-i., Hiraoka, M., 2000. Municipal solid waste incinerator residue recycling by
581 thermal processes. *Waste Manage.* 20, 249-258.

582 Savelainen, J., Isaksson, J., 2015. Kymijärvi II plant: High-efficiency use of SRF in
583 power production through gasification, Report by Lahti Energy Ltd and Metso
584 Power Oy, Finland.

585 Stein, W., Tobiasen, L., March 2004. Review of Small Scale Waste Conversion
586 Systems. IEA Bioenergy Agreement - Task 36, Work Topic 4.

587 Tanigaki, N., Manako, K., Osada, M., 2012. Co-gasification of municipal solid waste
588 and material recovery in a large-scale gasification and melting system. *Waste*
589 *Manage.* 32, 667-675.

590 The Commission of the European Communities, 2007. Commission Directive
591 2007/76/EC of 20 December 2007, amending Council Directive 91/414/EEC to
592 include fludioxonil, clomazone and prosulfocarb as active substances. Office
593 Journal of the European Union.

594 Turconi, R., Butera, S., Boldrin, A., Grosso, M., Rigamonti, L., Astrup, T., 2011. Life

595 cycle assessment of waste incineration in Denmark and Italy using two LCA
 596 models. *Waste Manage. Res.* 29, Supplement 78-90.

597 Van de Velden, M., Baeyens, J., Brems, A., Janssens, B., Dewil, R., 2010.
 598 Fundamentals, kinetics and endothermicity of the biomass pyrolysis reaction.
 599 *Renew. Energ.* 35, 232-242.

600 Vergara, S.E., Tchobanoglous, G., 2012. Municipal solid waste and the environment: a
 601 global perspective. *Annu. Rev. Env. Resour.* 37, 277-309.

602 Wäger, P.A., Hischer, R., 2015. Life cycle assessment of post-consumer plastics
 603 production from waste electrical and electronic equipment (WEEE) treatment
 604 residues in a Central European plastics recycling plant. *Sci. Total. Environ.* 529,
 605 158-167.

606 Wang, H., Wang, L., Shahbazi, A., 2015. Life cycle assessment of fast pyrolysis of
 607 municipal solid waste in North Carolina of USA. *J. Clean Prod.* 87, 511-519.

608 Wenzel, H., Hauschild, M.Z., Alting, L., 1997. *Environmental Assessment of Products*,
 609 Volume 1: Methodology, tools and case studies in product development.
 610 Chapman and Hall, London, UK.

611 Wood, S., Fanning, M., Venn, M., Whiting, K., 2013. Review of state-of-the-art
 612 waste-to-energy technologies. London (UK). Stage two, Case Studies, 9-30.

613 Yassin, L., Lettieri, P., Simons, S.J., Germanà, A., 2009. Techno-economic
 614 performance of energy-from-waste fluidized bed combustion and gasification
 615 processes in the UK context. *Chem. Eng. J.* 146, 315-327.

616 Yi, Q., Feng, J., Lu, B., Deng, J., Yu, C., Li, W., 2013. Energy evaluation for lignite

617 pyrolysis by solid heat carrier coupled with gasification. *Energ. Fuel.* 27,
618 4523-4533.

619 Zhang, Y., Li, B., Li, H., Liu, H., 2011. Thermodynamic evaluation of biomass
620 gasification with air in autothermal gasifiers. *Thermochim. Acta* 519, 65-71.

621 Zhao, W., van der Voet, E., Zhang, Y., Huppes, G., 2009. Life cycle assessment of
622 municipal solid waste management with regard to greenhouse gas emissions:
623 Case study of Tianjin, China. *Sci. Total. Environ.* 407, 1517-1526.

624

Tables

Table 1

Characteristics of the MSW as received at the plant.

Characteristics (wt. %, as received basis)	
C	25
H	4
N	0.84
S	0.13
O (by difference)	12
Moisture	34
Ash	24
Lower heating value, MJ/kg	9.8

Table 2

List of emission factors used in theoretical analysis and case studies of commercial WtE plants (Unit: mg/Nm³).

	Theoretical analysis ^a				Commercial WtE plants ^b			
	Incinerator	Gas boiler-steam	Gas turbine/ turbine/	Internal combustion	Incineration	Pyrolysis	Gasification	Gasification- melting
		turbine	CC	engine				
CO	50	50	100	100	5.5	10	2	6.2
SO ₂	50	35	15	15	0.44	8	7	3.3
NO _x	200	200	120	100	41.4	166.9	161	20.9
HCl	10	10	0	0	1.9	5.1	1	3.7
PM	10	5	0	0	0.09	1.4	2	1
PCDD/Fs (ng-TEQ/m ³)	0.1	0.1	0.1	0.1	0.005	0.001	0.002	0.006
Hg	0.05	0.05	0.05	0.05	0.002	0.011	0.0001	0.02
Cd	0.05	0.05	0.05	0.05	0.001	0.006	n.a. ^c	n.a. ^c

^a MSW incineration accords with the Directive 2007/76/EC; while pyrolysis and gasification plants meet the limits of the Directive 2010/75/EU with some adaptations (see detailed assumptions in Supplementary Material).

^b Data based on four commercial operated WtE plants (see plant information and data source in Supplementary Material).

^c Data not available.

Table 3

Sensitivity analysis by changing of the plant efficiency by $\pm 10\%$, based on the data from theoretical analysis.

	Environmental impacts change ^a (%)						
	S1	S2	S3	S4	S5	S6	S7
GW	± 50.2	± 19.2	± 24.9	± 20.4	± 26.9	± 110.9	± 18.3
AC	± 19.2	± 23.5	± 17.8	± 18.4	± 20.8	± 12.7	± 14.0
TE	± 9.8	± 12.1	± 12.2	± 11.9	± 15.5	± 23.9	± 34.6
POFh	± 14.6	± 17.8	± 665.5	± 480.2	± 24.5	± 22.0	± 32.2
HTa	± 2.8	± 3.6	± 2.0	± 1.9	± 4.1	± 11.9	± 6.4
HTs	± 0.4	± 0.3	± 0.2	± 0.2	± 0.3	± 0.4	± 0.3
ETs	± 0.1	± 0.1	± 0.1	± 0.1	± 0.1	± 0.2	± 0.1

^a Results represent a percentage increase or decrease of the environmental impacts in the base case scenarios.

Figure captions

Fig. 1. System boundaries of the study.

Fig. 2. Normalized environmental impacts of different systems based on the theoretical analysis: (a) non-toxic impacts; (b) toxic impacts.

Fig. 3. Contributional analysis for each environmental impact based on the theoretical analysis.

Fig. 4. Environmental impacts of different systems based on case studies of the selected commercial WtE plants: (a) non-toxic impacts; (b) toxic impacts.

Fig. 5. Sensitivity analysis by alternative utilization of the pyrolysis char, based on the data from theoretical analysis. Corner mark “LA” stands for land application as soil amendment.

(a) Non-toxic impacts

(b) Toxic impacts

Global warming (GW)

Acidification (AC)

Terrestrial eutrophication (TE)

Photochemical ozone formation - human health (POFh)

Human toxicity via air (HTa)

Human toxicity via solid (HTs)

Ecotoxicity via solid (ETs)

(a) Non-toxic impacts

(b) Toxic impacts