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► To cite this version:

Jun Dong, Yuanjun Tang, Ange Nzihou, Yong Chi, Elsa Weiss-Hortala, et al.. Life cycle assessment of pyrolysis, gasification and incineration waste-to-energy technologies: theoretical analysis and case study of commercial plants. *Science of the Total Environment*, 2018, 626, pp.744-753. 10.1016/j.scitotenv.2018.01.151 . hal-01712379v2

HAL Id: hal-01712379

<https://imt-mines-albi.hal.science/hal-01712379v2>

Submitted on 8 May 2018

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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

1 **Main text**

2 **1. Introduction**

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

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

23 device such as a gas turbine/combined cycle (gas turbine/CC) or an internal
24 combustion engine (Arena, 2012). Even if in a steam cycle plant, the limitation of
25 efficiency could be overcome by adding gas pre-treatment before it goes into the
26 burner, to allow the removal of HCl and an improvement in steam temperature of
27 520-540 °C (Belgiorno et al., 2003). Besides, pyrolysis and gasification have the
28 potential to diminish PCDD/Fs (Noma et al., 2012), thus reducing the total generation
29 of pollutants if the downstream syngas oxidization is processed efficiently. However,
30 using of the newly developed WtE options does not automatically guarantee the total
31 sustainability of the whole multi-stage thermal conversion and energy utilization
32 chain (Ning et al., 2013; Wang et al., 2015). The “raw” syngas, which contains a
33 variety of contaminants such as H₂S, tar, NH₃ and particulate matter (PM), needs to be
34 purified to meet the stringent requirement of entering an engine (Wood et al., 2013).
35 The configuration of different energy cycles downstream may also influence the
36 overall environmental effects: the consumptions and losses of gasification and syngas
37 clean-up may cause the overall energy efficiency be close or lower to incineration. It
38 is not a simple procedure to select an optimal WtE technology. A comprehensive
39 assessment of different WtE process configurations is necessary to understand if
40 pyrolysis/gasification-based WtE may become potential alternative or improvement
41 for the current incineration.

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

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

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

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

73

74 **2. Methodology**

75 2.1. System definition

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

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

95 The functional unit is set at one ton of MSW as received at the plant. Upstream
96 production of fuels and materials including diesel, electricity, lime, etc. is considered
97 as the ‘cradle to grave’ type of calculation. The benefits from useful co-products, such
98 as electricity and heat, are allocated by system expansion. The recovered electricity is
99 assumed to substitute that provided by the “energy mix” of a specific region, here the
100 European average (42.7% fossil fuels, 26.5% nuclear, 30.0% renewable energies,
101 0.7% waste and 0.1% other in 2015) is selected (Eurostat). The produced heat
102 displaces an equal amount of heat generated by “heat mix”, of which the heat
103 production data based on European average is again used (69.3% fossil fuels, 0.2%
104 nuclear, 22.9% renewable energies, 4.8% waste and 2.7% other in 2015) (Eurostat).
105 The database Gabi 7.0 provides the remaining, mainly indirect burdens, of the
106 background system.

107

108 2.2. MSW characteristics

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

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

115

116 2.3. Data source for theoretical analysis

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

124

125 2.3.1. MSW pre-treatment

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

133 2009). For systems using gas turbine/CC and internal combustion engine (S3, S4, S6
134 and S7), the heat derives from the syngas purification unit which recovers the sensible
135 heat of the hot syngas during cooling. For S2 and S5, the heat is supplied by the hot
136 flue gas.

137

138 2.3.2. Thermal conversion (pyrolysis, gasification and direct incineration)

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

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

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

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

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

174

175 2.3.3. Energy utilization cycles

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

177 final energy utilization is operated on-site. The electrical efficiency of the incineration
178 plant is set at 22.5% (Arena, 2012; Morris and Waldheim, 1998), which represents an
179 average of the modern dedicated waste combustion systems. For pyrolysis and
180 gasification systems, steam cycle is the simplest option because the hot syngas could
181 undergo combustion in the gas boiler without purification. A higher efficiency can be
182 achieved (set at 27.8% in this study), since the homogenous and gas-phase
183 combustion is more controllable and effective (Consonni and Viganò, 2012). The
184 syngas can also be burned in a gas turbine/CC or an internal combustion engine.
185 Potentially, the electrical efficiencies would be higher (set at 35.5% and 25.0% for gas
186 turbine/CC and engine, respectively (Belgiorno et al., 2003; Morris and Waldheim,
187 1998)). However, the syngas needs to be cooled and purified to meet the stringent
188 inlet gas quality requirement. To ensure the transparency of the data, the values of
189 plant efficiencies are determined by extensively searching and comparing with similar
190 set-up in the literature and reports (see details in **Table S2**). Additionally, a range of
191 variations of each plant efficiency will be discussed in the sensitivity analysis. For all
192 the systems analyzed, 20% of the generated electricity is assumed to be self-consumed
193 in the plant, with the remaining 80% sent to the power grid.

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

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

207

208 2.3.4. Emissions at the stack

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

220

221 2.3.5. Ash and air pollution control residues management

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

231

232 2.4. Data source for commercial operation WtE plants

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

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

243 moving grate combustion lines, having a treatment capacity of 450,000 t/a. MSW is
244 incinerated at 850 °C to produce electricity and district heating at an efficiency of
245 24% (net) and 6%, respectively (Turconi et al., 2011). The flue gas cleaning includes
246 electrostatic precipitator, acid gas neutralization (NaHCO₃ injection), fabric filter and
247 a SCR unit for NO_x abatement (Amsa, April, 2008). After combustion, metals are
248 sorted from the bottom ash and recycled. 88% of the bottom ash is utilized in road
249 construction, while the remaining fraction is landfilled and the APC residues are
250 safely disposed.

251 ● **Pyrolysis plant (C2)**: The selected plant, located in Hamm, Germany, has a
252 capacity of 100,000 t/a, although it is no longer in operation after the chimney
253 collapse in 2009. The pyrolysis process belongs to the RWE-ConTherm[®] technology
254 (DGEEngineering - The rotary kiln engineers, July 2009a). After shredded to 200 mm,
255 the MSW is decomposed in the absence of air in a rotary kiln at 500 °C with a
256 residence time of 1 hour, using natural gas as the heating source. The products, hot
257 syngas and char, are incinerated in the boiler of a coal-fired plant for electricity
258 production. The residues are considered to be landfilled and the metals are recycled.
259 The plant electricity efficiency (gross) is around 22% (Stein and Tobiasen, March
260 2004).

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

265 850-900 °C. The syngas generated undergoes cooling at 400 °C to remove heavy
266 metals and PM. The cleaned syngas enables a more efficient heat recovery boiler at
267 121 bar and superheated steam at 540 °C. The plant attains final 27% of electricity
268 efficiency (net) and 61% of heat efficiency (Savelainen and Isaksson, 2015). The flue
269 gas cleaning system consists of a bag house filter with additive injections (NaHCO₃
270 and activated carbon) and a SCR for NO_x reduction. From the plant outlet, the bottom
271 ash is removed to landfill disposal and the APC residues are safety disposed.

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

284

285 2.5. Life cycle inventory

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

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

292

293 2.6. Life cycle impact assessment

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

301

302 **3. Results**

303 3.1. Theoretical analysis results

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

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

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

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

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

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

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

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

361

362 3.2. Case studies for commercial operation WtE plants

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

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

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

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

397 materials.

398

399 3.3. Sensitivity analysis

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

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

410 For pyrolysis systems (S2-S4), the sensitivity analysis considers also the case
411 where the char is used as soil amendment. In such case, the pyrolysis char is
412 considered to have two additional merits (Harder and Forton, 2007; Roberts et al.,
413 2009): substitution of fertilizer (N, P and K) and carbon sequestration. Key
414 assumptions and calculations are presented in **Table S10**. **Fig. 5** indicates that this
415 assumption has exhibited an obvious reduction on the majority of impacts except for
416 HTs and ETs. The benefit is dominantly attributed to the reduced airborne emissions
417 from char combustion, together with a small portion of avoided emissions from
418 fertilizer substitution and carbon sequestration. However, a non-negligible increase of

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

422

423 **4. Discussions**

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

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

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

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

448 Those potential areas of development could include:

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

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

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

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

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

- 466 1. Syngas purification and use in higher energy efficiency equipment such as a
467 dedicated gas turbine/CC.
- 468 2. Further processing of syngas to be used as chemical feedstock, liquid fuels, etc.

469

470 **Acknowledgments**

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

474

475 **Appendix A. Supplementary data**

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

479

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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	Gas	Internal	Incineration	Pyrolysis	Gasification	Gasification- melting
		boiler-steam turbine	turbine/ CC	combustion 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

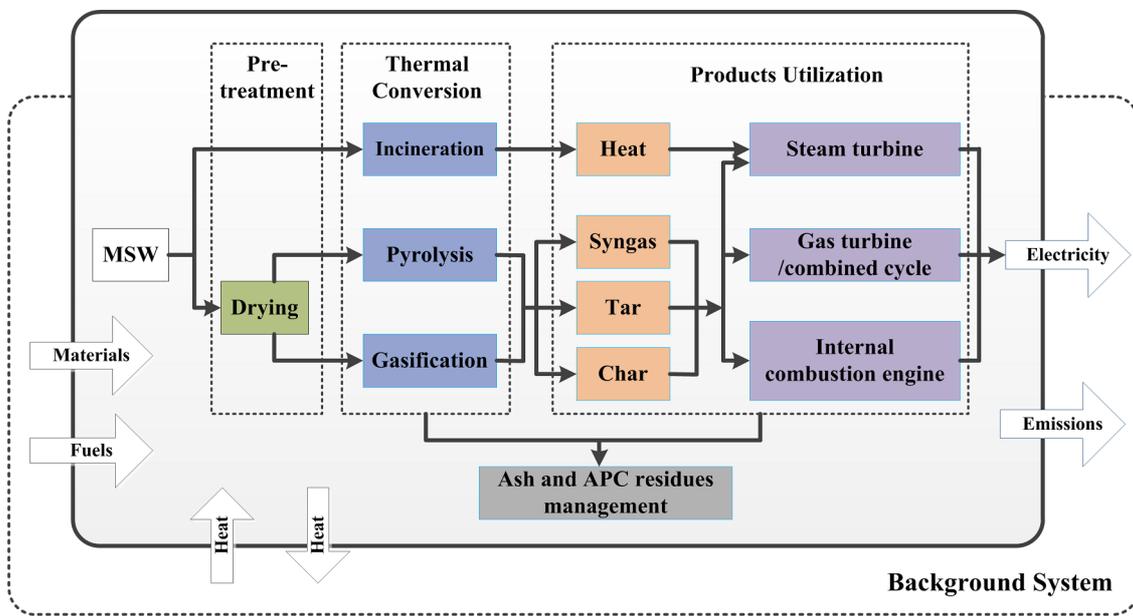
Fig. 1. System boundaries of the study.

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

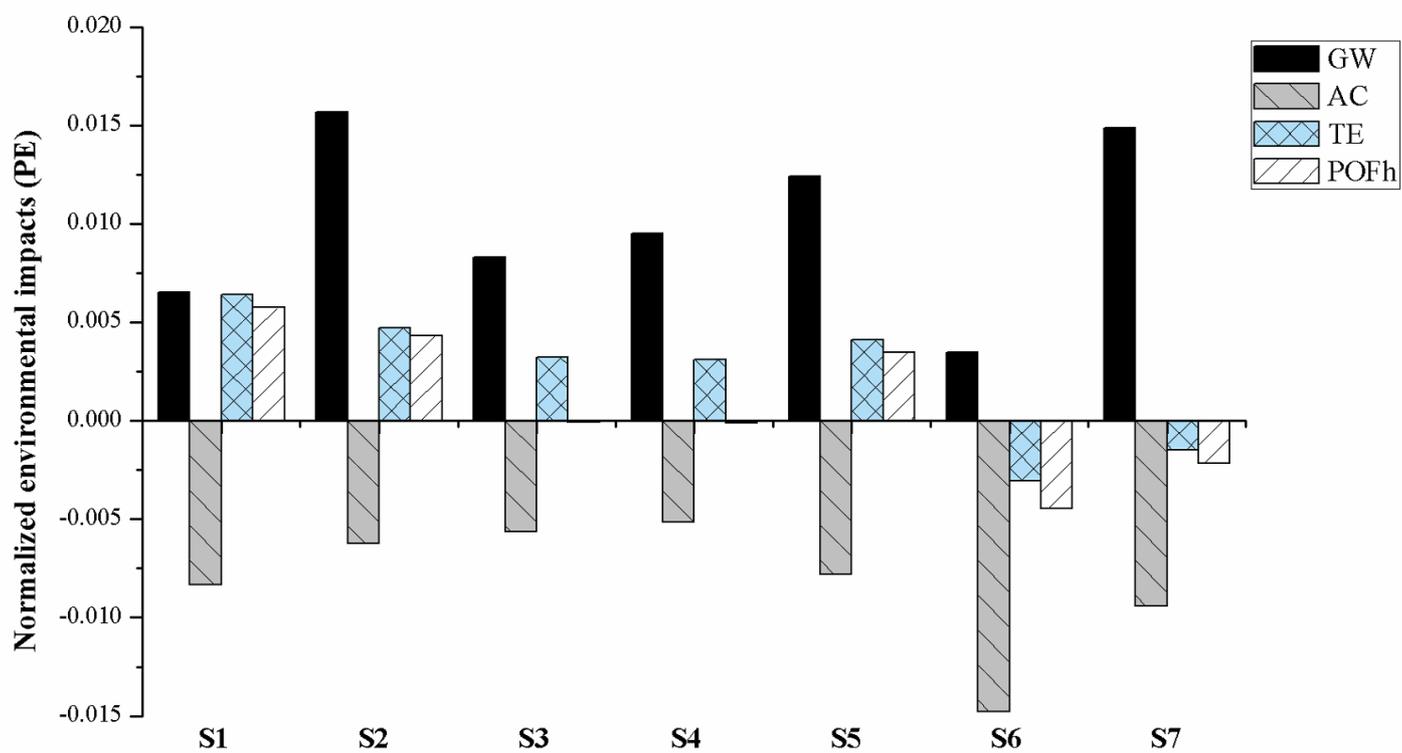
Fig. 3. Contributonal analysis for each environmental impact based on the theoretical analysis.

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

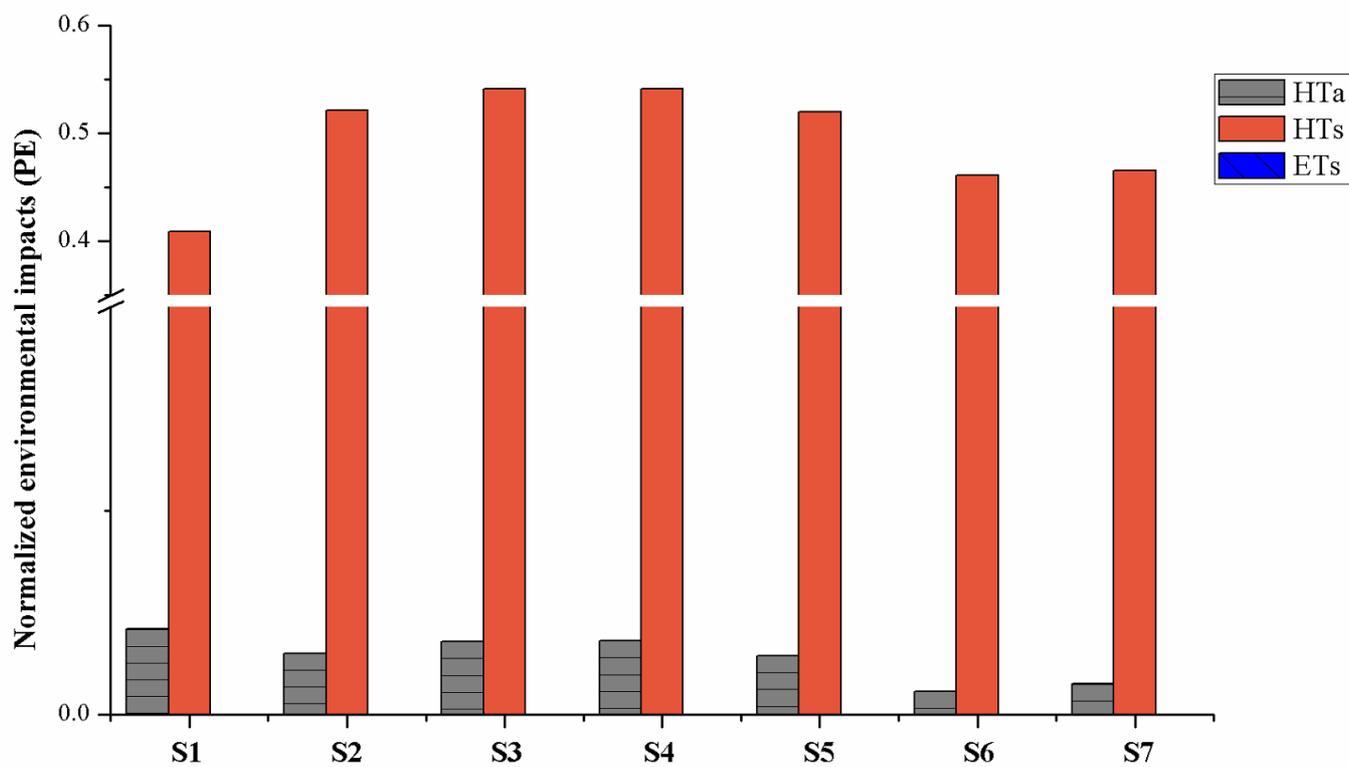
Fig. 5. Sensitivity analysis by alternative utilization of the pyrolysis char, based on the data from theoretical analysis. Corner mark “LA” stands for land application as soil amendment.



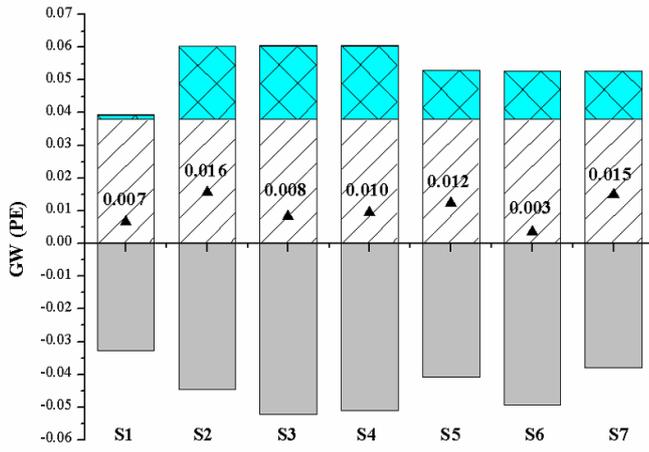
(a) Non-toxic impacts



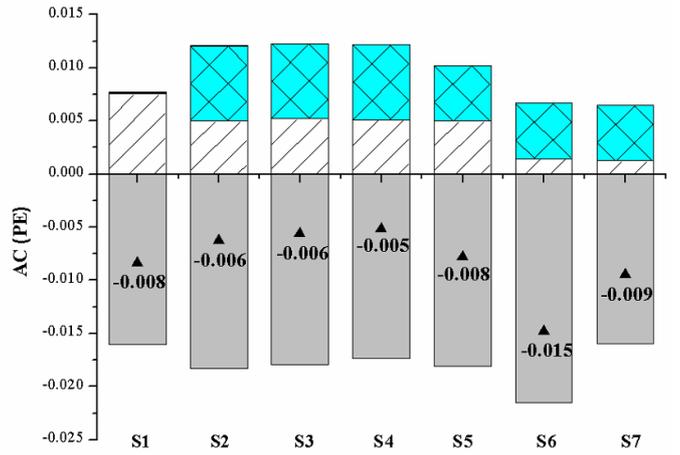
(b) Toxic impacts



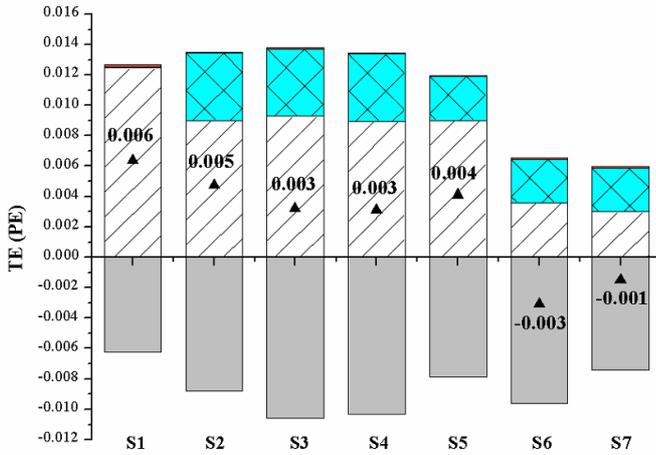
Global warming (GW)



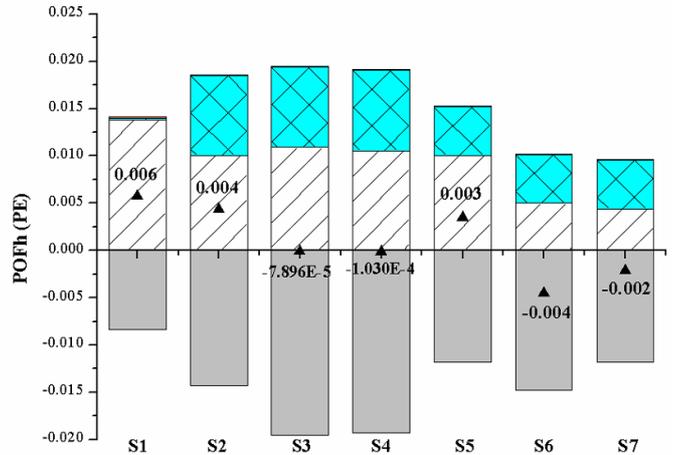
Acidification (AC)



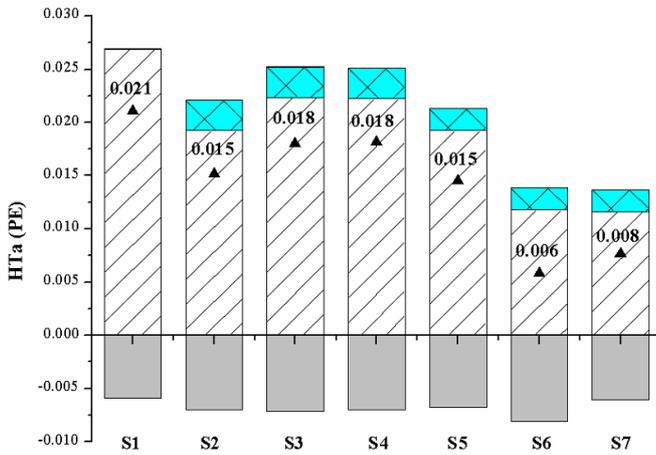
Terrestrial eutrophication (TE)



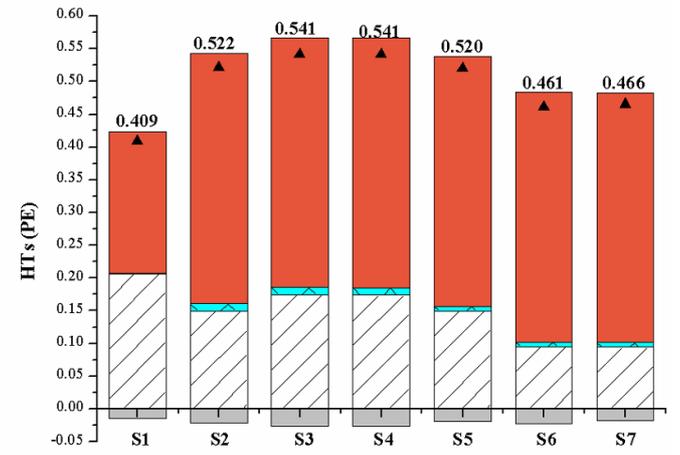
Photochemical ozone formation - human health (POFh)



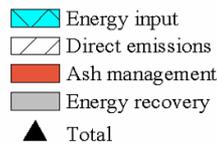
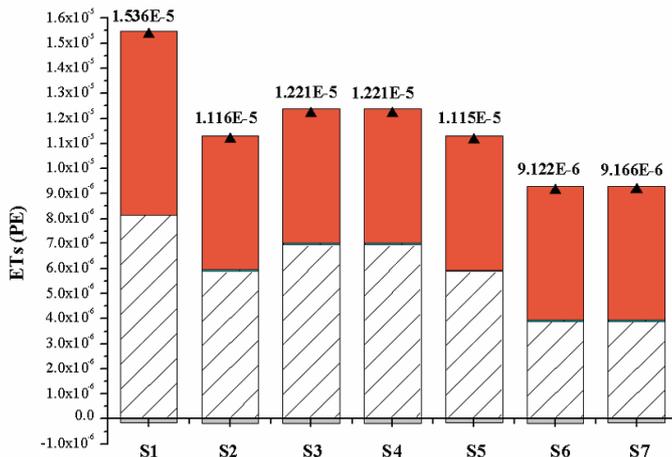
Human toxicity via air (HTa)



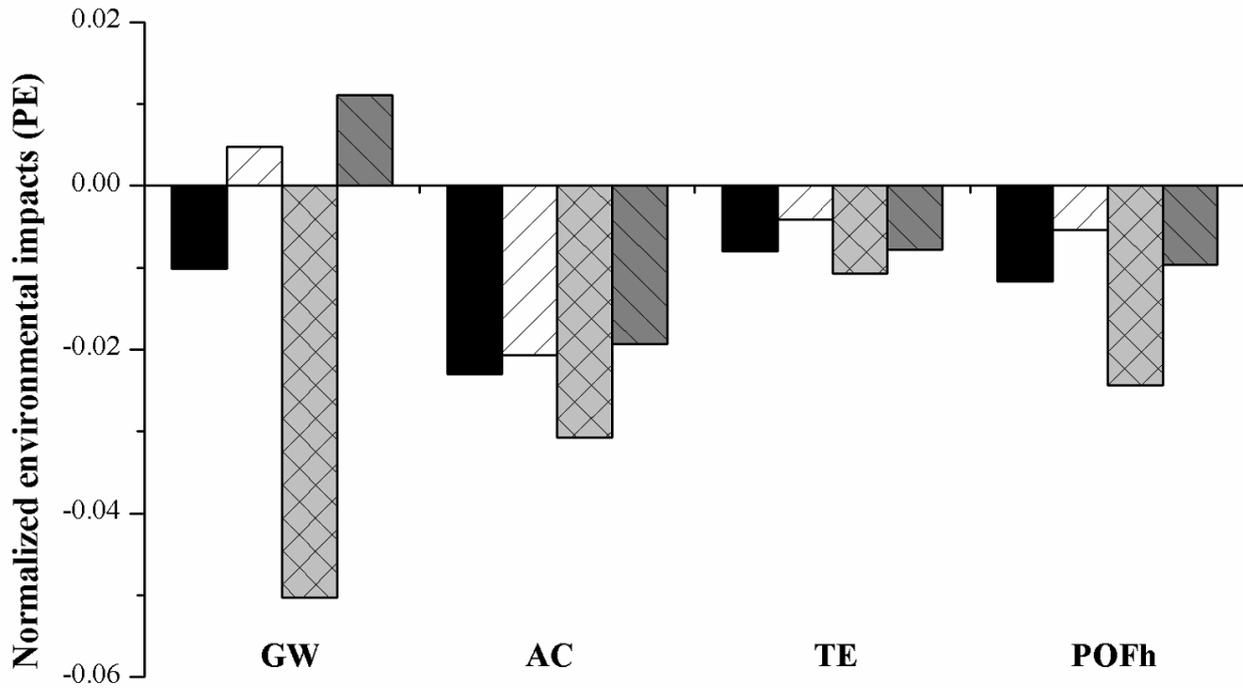
Human toxicity via solid (HTs)



Ecotoxicity via solid (ETs)



(a) Non-toxic impacts



(b) Toxic impacts

