

Sustainability assessment of furnace versus in-duct sorbent injection to retrofit waste-to-energy dry flue gas treatment lines

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To guarantee a safe compliance with the ambitious targets of acid gas (HCl, SO₂) removal set by recent European regulations, existing waste-to-energy (WtE) plants equipped with single-stage acid gas treatment systems might take into consideration the installation of an additional treatment stage as retrofit. Multi-stage treatment systems can achieve higher acid gas removal efficiency and flexibility to cope with highly variable inlet pollutant concentrations, resulting from variability in waste composition.

From a sustainability viewpoint, the choice of the most suited retrofitting option has to be driven not only by considerations on performance at the WtE plant but also by considerations on the indirect environmental burdens related to the material and energy consumption and waste generation of the selected options.

The present study analyses the full environmental profile of two competitive dry retrofitting solutions (the low-temperature injection of hydrated lime and the high-temperature injection of calcined dolomite) by considering their life cycle impacts associated with the supply of reactants and the disposal of process residues. Modelling of the material balance of the alternatives as a function of pollutant load in the inlet flue gas allowed estimating the effect of flue gas composition and mode of operation of the two-stage system on the overall environmental impact. Both retrofitting options were found to reduce the life cycle impacts of the base case, but the use of hydrated lime showed the greatest potential. The generation of process residues constitutes a relevant fraction of the overall impacts. An alternative scenario for the management of process residues was evaluated to quantify the reduction of impact achievable with a commercially available valorisation route.

1. Introduction

The air emission of a wide range of pollutants, potentially harmful for both human health and ecosystem integrity, is typically considered as the main drawback of the thermal valorisation of waste in waste-to-energy (WtE) plants (Cole-Hunter et al., 2020). To address this issue, which also generates frequent opposition to the construction and operation of incinerators by local communities (Zheng et al., 2021), the WtE sector is generally subject to more stringent emission limits than other industrial sectors (Dal Pozzo et al., 2021). In Europe, the latest revision of the *Best Available Techniques Reference Document for Waste Incineration* (BREF WI, Neuwahl et al., 2019) is setting ambitious targets for emission reduction that existing WtE plants need to meet in order to renew their environmental permits within 2024.

In particular, a required area of improvement is the control of the emissions of acid compounds, namely HCl and SO₂, fuel-related pollutants that arise from the S and Cl content of the waste burnt in the plant (Zhang et al., 2019). As recent surveys pointed out (Beylot et al., 2018; Dal Pozzo et al., 2018a), the most common method for acid gas removal is their neutralisation by in-duct injection of dry powdered alkaline sorbents and the subsequent filtration of solid reaction products, with sodium bicarbonate being the prevalent choice as sorbent. Existing WtE plants equipped with such a dry injection system can adapt their operation to newer, stricter emission limits by significantly increasing the feed rate of sorbent.

Alternatively, they can install an additional stage of acid gas treatment, thus realizing a two-stage abatement system. The resulting redundancy of acid gas removal capacity guarantees high performances and increased flexibility in coping with sudden peaks of inlet HCl or SO₂ load (De Greef et al., 2013). In addition, it has been demonstrated that properly operated two-stage systems can achieve significant savings in the cost of operation compared to single-stage bicarbonate-fed systems (Dal Pozzo et al., 2016).

For these reasons, in recent years two approaches for the realisation of a two-stage acid gas removal systems have emerged in the WtE market: i) the addition of a high-temperature furnace sorbent injection of slaked dolomite, and ii) the addition of a low-temperature in-duct sorbent injection of hydrated lime. In both cases, the second stage of acid gas treatment is a dry sorbent injection of sodium bicarbonate. As an example of the recent diffusion of these configurations, Fig. 1 shows their distribution among Italian WtE installations.

Nowadays, a holistic approach to sustainability demands that the evaluation of the environmental performance of flue gas cleaning techniques be not limited to their air emissions. Also the indirect environmental burdens related to the material and energy consumption of the technological alternatives, and the generation of possible solid residues, need to be considered. The present study aims at offering an analysis of such indirect environmental burdens for the two considered retrofitting options by adopting a life cycle approach, encompassing the supply chain of reactants and the fate of solid residues generated by the flue gas treatment process.

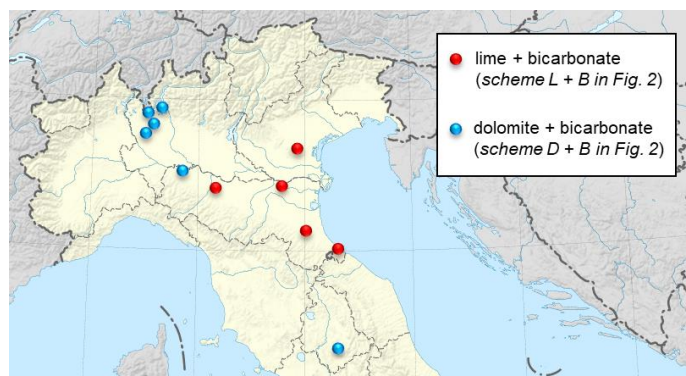


Figure 1: WtE plants in Italy currently adopting one of the two retrofitting options analysed in the study.

2. Reference technologies

Fig. 2 illustrates the layout of the two retrofitting solutions analysed in the study for the conversion of a conventional single-stage acid gas removal system to a two-stage system.

The conventional single-stage system consists in the in-duct injection of sodium bicarbonate, NaHCO_3 . At the typical temperature of injection ($> 150\text{ }^\circ\text{C}$), bicarbonate decomposes to carbonate, which in turn reacts with the acid pollutants (see Table 1). The reactions take place both in the entrained flow in the duct and at the surface of the filter bags in the subsequent fabric filter.

In the configuration L+B (Fig. 2a), the retrofit consists in the introduction of an additional pre-treatment stage of reaction and filtration, based on the in-duct injection of hydrated lime, $\text{Ca}(\text{OH})_2$. Conversely, in the configuration D+B (Fig. 2b), pre-treatment takes place directly in the combustion chamber by injection of calcined dolomite, $\text{Ca}(\text{OH})_2\cdot\text{MgO}$. As shown in Table 1, the sets of reactions with acid compounds are similar and driven by Ca in both cases, as in the dolomitic sorbent MgO acts only as structural agent to avoid high-temperature sintering (Biganzoli et al., 2015a).

The products of the acid gas removal processes are solid residues. They are separated from the gas stream together with combustion fly ash and typically have adsorbed also trace pollutants (e.g. Hg and PCDD/F). As such, they are considered hazardous waste and to-date destined to landfilling (Dal Pozzo et al., 2020c; Kameda et al., 2020), although several valorisation routes are currently under research (Margallo et al., 2018).

Only bicarbonate-based residues might be sent to recycling plants that regenerate fresh bicarbonate (Biganzoli et al., 2015b), if the residues are collected separately from fly ash, i.e. a dedusting equipment is installed before bicarbonate injection.

3. Methodology

3.1 Case study

The case study for the comparison of the environmental impacts of the two alternatives was set up as follows. The functional unit for the comparison was 1 year of operation at a medium-sized WtE installation (400 t/d of treated waste, $100,000\text{ Nm}^3/\text{h}$ of generated flue gas, 7200 h/y of operation). To take into account the effect of the typically high variability of HCl and SO_2 concentration in WtE plants, three distinct subcases of inlet flue gas composition entering the treatment system were defined (Table 2). It was imposed as process specification a setpoint of HCl emission equal to $0.5\text{ mg}/\text{Nm}^3$, guaranteeing a safety margin with respect to the emission limit values recommended by BREF WI (Neuwahl et al., 2019).

Table 1: Reactions occurring in the acid gas removal process.

Ca(OH)_2 reactions	$\text{Ca(OH)}_2 \cdot \text{MgO}$ reactions	NaHCO_3 reactions
$\text{Ca(OH)}_2 + 2 \text{HCl} \rightarrow \text{CaCl}_2 + 2 \text{H}_2\text{O}$ (R1)	$\text{Ca(OH)}_2 \cdot \text{MgO} \rightarrow \text{CaO} \cdot \text{MgO} + \text{H}_2\text{O}$ (R3)	$2 \text{NaHCO}_3 \rightarrow \text{Na}_2\text{CO}_3 + \text{CO}_2 + \text{H}_2\text{O}$ (R6)
$\text{Ca(OH)}_2 + \text{SO}_2 + \frac{1}{2} \text{O}_2 \rightarrow \text{CaSO}_4 + \text{H}_2\text{O}$ (R2)	$\text{CaO} + 2 \text{HCl} \rightarrow \text{CaCl}_2 + \text{H}_2\text{O}$ (R4)	$\text{Na}_2\text{CO}_3 + 2 \text{HCl} \rightarrow 2 \text{NaCl} + \text{CO}_2 + \text{H}_2\text{O}$ (R7)
	$\text{CaO} + \text{SO}_2 + \frac{1}{2} \text{O}_2 \rightarrow \text{CaSO}_4$ (R5)	$\text{Na}_2\text{CO}_3 + \text{SO}_2 + \frac{1}{2} \text{O}_2 \rightarrow \text{Na}_2\text{SO}_4 + \text{CO}_2$ (R8)

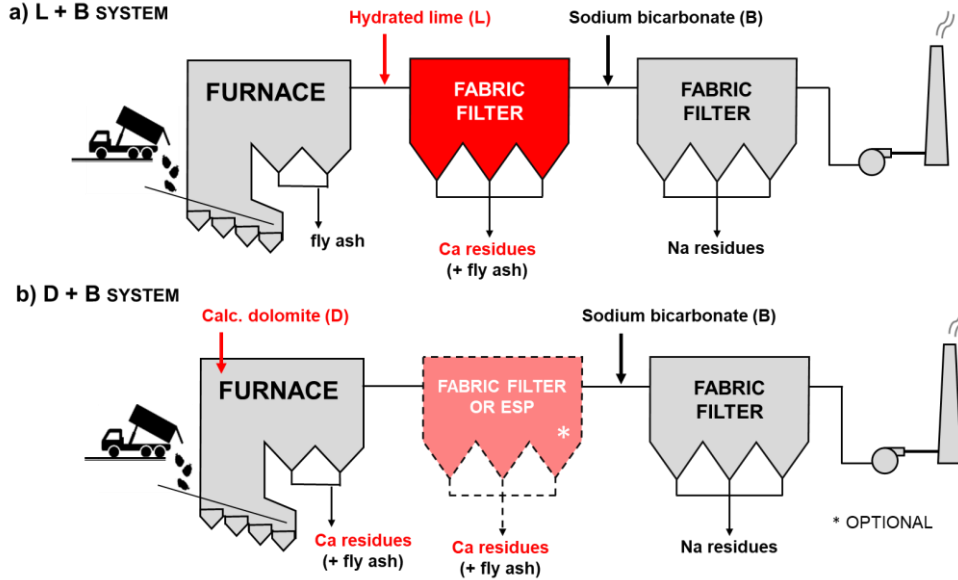


Figure 2: Schemes of two-stage acid gas removal systems obtained by adding a) a low-temperature hydrated lime injection and b) a high-temperature calcined dolomite injection to an existing single stage bicarbonate-fed treatment system. The modifications with respect to the original single stage configuration are highlighted in red.

Table 2: Concentration of acid compounds (in mg/Nm^3) in the 3 cases of inlet flue gas composition.

Acid compound	Case 1	Case 2	Case 3
HCl	1000	600	1000
SO_2	100	100	300

3.2 Modelling of the systems and the related supply and disposal chain

To evaluate the overall environmental impacts of the alternatives, the analysis encompassed the direct impacts generated by the treatment systems (viz. the residual emission of acid pollutants and the generation of CO_2 by NaHCO_3 decomposition) and the indirect impacts generated along the supply chains of reactants and the disposal routes of process residues.

Starting point for the analysis was the quantification of the feed rate of reactants required to obtain a given acid gas removal efficiency and the related generation rate of solid residues by means of mass balances for the acid gas removal processes. A simple, yet widely adopted model (Dal Pozzo et al., 2016) was used to correlate the conversion of the acid pollutants i by the reactant j :

$$X_{i,j} = \frac{rs_{i,j}^{n_{i,j}} - rs_{i,j}}{rs_{i,j}^{n_{i,j}} - 1} \quad (1)$$

where $rs_{i,j}$ is the ratio between the actual feed rate of the solid reactant j and the stoichiometric rate theoretically required for the total conversion of all the acid gases (see reactions in Table 1) and $n_{i,j}$ are empirical parameters, tuned on actual operational data. Details on the determination of $n_{i,j}$ for Ca(OH)_2 and NaHCO_3 via historical process analysis and calcined dolomite via dedicated test runs can be found elsewhere (Dal Pozzo et al., 2018b, 2020). Here, it can be recalled that Ca(OH)_2 is markedly less reactive than NaHCO_3 towards both HCl and SO_2 , owing to diffusional limitations hindering its reactivity (Antonioni et al., 2016), while calcined dolomite exhibits an accentuated affinity towards SO_2 than HCl (Biganzoli et al., 2015a).

Whereas the model was used to characterise the main foreground process (i.e. the operation of the treatment system), inventories for the other processes were reconstructed from scientific and technical literature:

- the production of $\text{Ca}(\text{OH})_2$ was retrieved from the ecoinvent LCI database;
- the production of NaHCO_3 was modelled as in Dong et al. (2020);
- the production of calcined dolomite was modelled as in Biganzoli et al. (2015b);
- the disposal of process residues after stabilisation with cement was modelled as in Margallo et al. (2018);
- the transportation phases were considered to be performed by lorry for distances of 150 km (lime supply, landfill disposal) or 300 km (bicarbonate supply), following the detailed analysis of available facilities in the Italian context by Dal Pozzo et al. (2017).

3.3 Impact assessment

A set of 5 problem-oriented impact indicators – namely: resource depletion (RD), global warming (GW), acidification (AC), smog formation (SF), particulate formation (PF) – was adopted to characterise the environmental impacts. Characterisation factors for all categories were retrieved from the CML-IA database (CML, 2016) and were expressed in terms of equivalent mass of a relevant reference substance.

4. Results and discussion

Being two-stage systems, the two alternatives of Fig. 2 present a degree of freedom in their utilisation: given a fixed overall acid gas removal efficiency as set in section 3.1, this overall target can be achieved by different repartitions of removal between the two stages. For the case 1 of flue gas composition, Fig. 3 explores how a different degree of utilisation of the two stages, expressed as ratio of HCl conversion in the 1st treatment stage to total HCl conversion ($X_{1^{\text{st}} \text{ stage}} / X_{\text{TOT}}$), influences the overall environmental impacts of the L+B and D+B systems. The scores in the five indicators are normalised to the impact of the single stage bicarbonate system before retrofitting, which corresponds to a use of the 1st treatment stage (lime or bicarbonate) equal to zero. It can be seen that in the L+B system the feed of $\text{Ca}(\text{OH})_2$ to obtain up to 80% of the overall HCl removal in the 1st treatment stage realises a significant reduction of impact compared to the single stage system. Conversely, the D+B system exhibits slightly lower impacts in all the categories compared to the single stage system only for HCl conversions in the 1st stage lower than 15%.

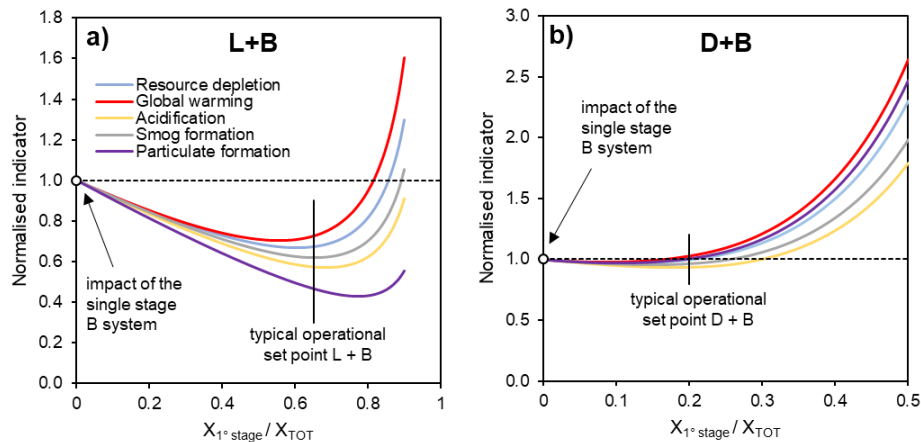


Figure 3: Evolution of the environmental impacts of acid gas removal as a function of HCl conversion in the 1st treatment stage: a) L+B system, b) D+B system. Flue gas composition: case 1.

For the following comparative analysis, a single operating point was selected for the two alternatives. For the L+B system it was selected the operation of the system with 65% HCl conversion in the 1st stage, which previous studies identified as the optimal operating point from an economic point of view (Dal Pozzo et al., 2016). Likewise, following examples of typical operation (Biganzoli et al., 2015a; Dal Pozzo et al., 2020), for the D+B system it was selected as preferred operating point a 20% HCl conversion in the 1st stage.

Table 3 summarises the impacts related to the selected operating points. The L+B option emerges as the preferable technology in all the categories, with impacts that ranges from 31% lower (GW indicator) to 57% lower (PF indicator) compared to D+B for the flue gas composition of case 1. In case 2 and 3, the absolute values of the indicators change significantly (e.g. the GW indicator for L+B is 37% lower in case 2 and 25% higher in case 3), as a demonstration of the importance of flue gas composition in determining the overall impact of acid gas removal, but the relative advantages of the L+B solution vs. D+B are confirmed.

Table 3: Indicators for L+B and D+B at their selected operating points for the 3 cases of flue gas composition.

Indicator	Unit	Case 1		Case 2		Case 3	
		L+B	D+B	L+B	D+B	L+B	D+B
RD	MJ/y	1.57×10^7	2.44×10^7	9.86×10^6	1.47×10^7	1.96×10^7	2.85×10^7
GW	kgCO _{2,eq} /y	3.02×10^6	4.40×10^6	1.90×10^6	2.66×10^6	3.76×10^6	5.20×10^6
AC	kgSO _{2,eq} /y	4.35×10^3	7.23×10^3	2.86×10^3	4.47×10^3	5.40×10^3	8.20×10^3
SF	kgethylene,eq/y	2.84×10^2	4.67×10^2	1.79×10^2	2.82×10^2	3.57×10^2	5.39×10^2
PF	disease inc./y	5.54×10^{-2}	1.28×10^{-1}	3.51×10^{-2}	7.71×10^{-2}	7.02×10^{-2}	1.50×10^{-1}

To offer a comparison of the presented results with relevant literature, the GW score of the D+B solution in case 2 (2.66×10^6 kgCO_{2,eq}/y) corresponds to 22 kgCO_{2,eq}/t_{waste}, a value that is in line with the 23 kgCO_{2,eq}/t_{waste} determined by Biganzoli et al. (2015b) under similar assumptions.

Fig. 4 clarifies the contribution of the single life cycle phases to the overall impacts of the two alternatives. The production of sodium bicarbonate, an energy-intensive process, and the disposal of process residues at landfills emerge as the main contributors to environmental impact in most categories. In addition, bicarbonate is responsible for direct impacts at the WtE plant for the GW category, owing to the CO₂ release associated with its decomposition.

It is interesting to notice that, in the acidification category, the impact related to the direct emission at the stack of the WtE plant of the residual HCl and SO₂ not captured by the acid gas removal system is only a marginal contribution (<5%) compared to the indirect impacts arising in the life cycle phases. Once again, this demonstrates the high performance level achieved by state-of-the-art WtE abatement technologies at plant and the current need to shift attention to the indirect life cycle impacts to obtain further environmental gains.

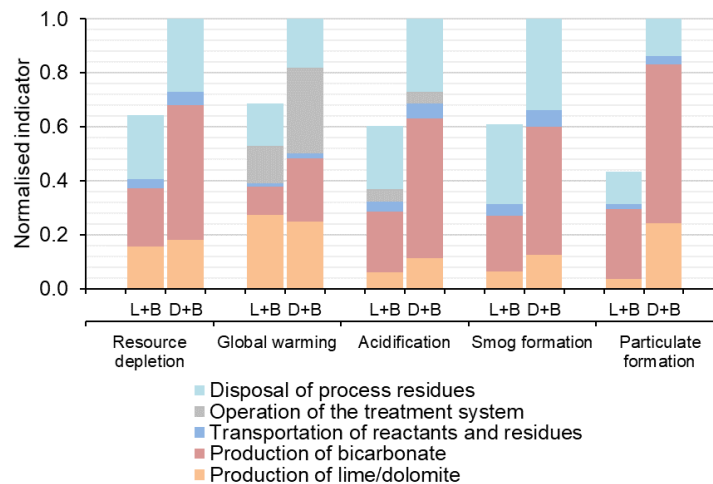


Figure 4: Indicators of the environmental impact of the two alternatives at their selected operating point (see Figure 3) and contribution of the single life cycle phases. Internal normalisation. Flue gas composition: case 1.

Table 4: Reduction of environmental impacts achievable by recycling the bicarbonate residues.

Indicator	Unit	Case 1 - recycling		Impact reduction	
		L+B	D+B	L+B	D+B
RD	MJ/y	1.38×10^7	2.01×10^7	12%	18%
GW	kgCO _{2,eq} /y	2.79×10^6	3.86×10^6	8%	12%
AC	kgSO _{2,eq} /y	3.77×10^3	5.93×10^3	13%	18%
SF	kgethylene,eq/y	2.39×10^2	3.65×10^2	16%	22%
PF	disease inc./y	4.33×10^{-2}	1.00×10^{-1}	22%	21%

As an example of actions that can be pursued to mitigate life cycle impacts, it is here explored the possibility to send the process residues of bicarbonate injection to recycling instead of landfilling. As mentioned in section 2, this option is technically feasible only if the bicarbonate-fed stage is preceded by an upstream dedusting equipment, as it is the case for both the L+B and D+B alternatives (see again Fig. 2). The recycling of bicarbonate residues was modelled as in Righi et al. (2018), also considering the avoided impacts of the supply of fresh reactants for bicarbonate production thanks to its regeneration from waste materials. Table 4 shows

that diverting the waste stream of bicarbonate residues from landfill to recycling can guarantee a sizeable impact reduction in all the categories. In particular, the advantages are greater for the D+B system, which produces more bicarbonate residues.

5. Conclusions

In the present work, a sustainability assessment of two alternative approaches for the retrofitting of WtE dry acid gas removal systems in the light of new environmental regulations is carried out. With state-of-the-art technologies reaching remarkably high removal efficiencies at the WtE plant, it is shown that indirect burdens arising in the life cycle of reactants and residues constitute a dominant share of the overall environmental impact. Among the considered options, the two-stage system based on lime and bicarbonate injection exhibits lower impacts than the dolomite + bicarbonate counterpart. In both cases, the production of sodium bicarbonate and the disposal of process residues are the main contributors of environmental impacts. The possibility to recycle residues for the regeneration of fresh bicarbonate can significantly reduce such burdens.

References

- Antonioni G., Dal Pozzo A., Guglielmi D., Tugnoli A., Cozzani V., 2016, Enhanced modelling of heterogeneous gas-solid reactions in acid gas removal dry processes, *Chem. Engin. Sci.* 148, 140-154.
- Beylot A., Hochar A., Michel P., Descat M., Menard Y., Villeneuve J., 2018, MSWI in France: An Overview of Air Pollution Control Techniques, Emissions, and Energy Efficiency, *J. Ind. Ecol.* 22, 1016-1026.
- Biganzoli L., Racanella G., Rigamonti L., Marras R., Grosso, M., 2015a, High temperature abatement of acid gases from waste incineration. Part I: Experimental tests in full scale plants, *Waste Manage.* 36, 98-105.
- Biganzoli L., Racanella G., Marras R., Rigamonti L., 2015b, High temperature abatement of acid gases from waste incineration. Part II: Comparative life cycle assessment study, *Waste Manage.* 35, 127-134.
- Cole-Hunter T., Johnston F.H., Marks G.B., (...), Cowie C.T., 2020, The health impacts of waste-to-energy emissions: a systematic review of the literature, *Environ. Res. Letters* 15, 123006.
- Dal Pozzo A., Antonioni G., Guglielmi D., Stramigioli C., Cozzani V., 2016, Comparison of alternative flue gas dry treatment technologies in waste-to-energy processes, *Waste Manage.* 51, 81-90.
- Dal Pozzo A., Guglielmi D., Antonioni G., Tugnoli A., 2017, Sustainability analysis of dry treatment technologies for acid gas removal in waste-to-energy plants, *J. Cleaner Prod.* 162, 1061-1074.
- Dal Pozzo A., Guglielmi D., Antonioni G., Tugnoli A., 2018a, Environmental and economic performance assessment of alternative acid gas removal technologies for waste-to-energy plants, *Sustainable Prod. Consumption* 16, 202– 215.
- Dal Pozzo A., Giannella M., Antonioni G., Cozzani V., 2018b, Optimization of the Economic and Environmental Profile of HCl Removal in a Municipal Solid Waste Incinerator through Historical Data Analysis, *Chem. Eng. Trans.* 67, 463-468.
- Dal Pozzo A., Armutlulu, A., Rekhtina M., Müller C.R., Cozzani V., 2018c, CO₂ Uptake Potential of Ca-Based Air Pollution Control Residues over Repeated Carbonation–Calcination Cycles, *Energy Fuels* 32, 5386-5395.
- Dal Pozzo A., Lazazzara L., Antonioni G., Cozzani V., 2020, Techno-economic performance of HCl and SO₂ removal in waste-to-energy plants by furnace direct sorbent injection, *J. Hazard. Mater.* 394, 122518.
- Dal Pozzo A., Muratori G., Antonioni G., Cozzani V., 2021, Economic and environmental benefits by improved process control strategies in HCl removal from waste-to-energy flue gas, *Waste Manage.* 125, 303-315.
- De Greef J., Villani K., Goethals J., (...), Vandecasteele C., 2013, Optimising energy recovery and use of chemicals, resources and materials in modern waste-to-energy plants, *Waste Manage.* 33, 2416-2424.
- Dong J., Jeswani H.K., Nzihou A., Azapagic A., 2020, The environmental cost of recovering energy from municipal solid waste, *Appl. Energy* 267, 114792.
- Kameda T., Tochinali M., Kumagai S., Yoshioka T., 2020, Treatment of HCl gas by cyclic use of Mg–Al layered double hydroxide intercalated with CO₃²⁻, *Atm. Pollut. Res.* 11, 290-295.
- Margallo M., Cobo S., Muñoz E., (...), Irabien A., 2018, Life cycle assessment of alternative processes to treat fly ash from waste incineration, *Chem. Eng. Trans.* 70, 883-888.
- Neuwahl F., Cusano G., Gomez Benavides J., Holbrook S., Roudier S., 2019. Best Available Techniques (BAT) Reference Document for Waste Incineration. EUR 29971 EN.
- Righi S., Baioli F., Dal Pozzo A., Tugnoli A., 2018, Integrating Life Cycle Assessment and Process Design Techniques for the Early Estimate of Energy and Material Consumption Data, *Energies* 11, 970.
- Zhang H., Yu S., Shao, L., He P., 2019, Estimating source strengths of HCl and SO₂ emissions in the flue gas from waste incineration, *J. Environ. Sci.*, 75, 370-377.
- Zheng J., Yu L., Ma G., Mi H., Jiao Y., 2021, Residents' acceptance towards waste-to-energy facilities: formation, diffusion and policy implications, *J. Cleaner Prod.* 287, 125560.