Indirect Environmental Benefits of a Variable Set-Point for HCl Emission in Waste-to-Energy Flue Gas Cleaning

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Hydrogen chloride (HCl) is a typical contaminant generated in waste combustion, whose emission is strictly regulated. In order to cope with severe limits issued by local authorities on the annual mass flow of HCl emitted, several European waste-to-energy plants perform at least a two-stage abatement of the pollutant and operate with a set-point for HCl concentration at stack well below the hourly emission limit imposed by the Industrial Emissions Directive. Even though the operating conditions in a WtE facility are highly variable (both in terms of HCl release from the burnt waste and of operating temperature of the HCl removal system), it is common industrial practice to keep both the set-point for HCl concentration at stack and the set-point for HCl concentration between the two abatement stages at a constant value. However, this might force the abatement system to operate far from its design conditions and from the optimal reactivity window of the adopted HCl removal agent, thus increasing the consumption rate of reactant and the consequent generation rate of process residues. Both the supply chain of reactants and the management of residues generate environmental impacts. The present work demonstrates that the adoption of variable values for both set-points can provide cost savings and indirect environmental benefits, thanks to the reduction of reactant consumption and residue generation. By considering as case study a WtE plant equipped with a two-stage dry HCl removal system, a year of operation of the plant was analysed. The operating costs and life cycle impacts generated by the actual operation of the HCl treatment system with constant set-points were compared with those generated if the plant had operated with variable set-points, simulated with a semi-empirical model for HCl removal tuned on historical process data from the plant. A 13 % reduction of the life cycle global warming impact and an 8 % reduction of the life cycle acidification impact, in addition to 10 % cost savings, were associated to the utilisation of variable set-points.

1. Introduction

WtE plants are a source of various airborne pollutants and, as such, are kept under close scrutiny by the surrounding populace and are subject to severe emission limits imposed in Europe by the Industrial Emissions Directive (IED). As contemplated by the directive, local authorities often impose additional limits on the annual mass flow of airborne pollutants emitted, frequently stricter than the IED daily emission limits.

Plant managers face this constraint by regulating the daily emission set-point for the target pollutant at a uniform value throughout the year, much lower than the IED daily emission limit. A typical case is the emission control for hydrogen chloride, a signature pollutant of waste incineration (Hasan et al., 2017): despite an IED daily emission limit of 10 mg/Nm^3, WtE plants operate by keeping a constant emission set-point of 4, 2 or even 1 mg/Nm^3, depending on the annual mass flow limit.

This solution might result sub-optimal on a broader environmental point of view. The HCl loads entering the treatment system are highly variable, due to the natural variability of the waste feedstock (Dal Pozzo et al., 2017a). By keeping constant the emission set-point, the system is thus asked to operate at a variable range of required HCl conversion, overstressed when the concentration of HCl in the flue gas is high and underused when it is lower. This might result in unnecessary extra consumption of reactants (generally, powdered sorbents; Yassin et al., 2007) and generation of process residues (the solid products of HCl neutralisation). In addition, the operating temperature at which the flue gas enters the HCl abatement system might also vary in time as a consequence of fouling in the upstream heat recovery section of the plant (Biganzoli et al., 2015) and the reactivity of some widely used reactants for HCl removal, such as Ca-based sorbents, is negatively affected

even by slight temperature increases (Weinell, 1992), *i.e.* more reactant is needed and more residues are generated in order to obtain the same removal efficiency. Both the supply chain of reactants and the management of residues generate indirect environmental impacts (Dal Pozzo et al., 2017b), hence these aspects have to be considered in a holistic approach to environmental protection.

The present study quantifies the potential indirect environmental gains that can be achieved by adopting a variable set-point for HCl emission instead of a constant one, keeping equal the annual mass flow limit of emitted pollutant. The case study is a reference WtE plant, facing a strict annual mass flow limit for HCl and equipped with a two-stage HCl dry treatment system. Process data from the plant were used to calibrate a semi-empirical model to quantify the sorbent feed rate needed to achieve a given HCl conversion and the related generation of solid residues. The environmental impacts associated with production, transportation and disposal/recycling of sorbents and residues were estimated adopting a life cycle perspective.

2. Methodology

2.1 Case study

The reference plant for the case study is a medium-sized WtE facility (45 MW_{th}) treating 16 t/h of waste and generating 100,000 Nm^3/h of flue gas. HCl abatement is performed by means of a two-stage dry treatment system, outlined in Figure 1. In this process scheme, the flue gas exiting the heat recovery section of the plant is first treated by the injection of hydrated lime, $Ca(OH)_2$. Calcium hydroxychloride (CaOHCl), the solid product of the neutralisation reaction between $Ca(OH)_2$ and HCl (Dal Pozzo et al., 2018), is collected downstream by a fabric filter, together with some unreacted lime and the calcium carbonate ($CaCO_3$) generated by the undesired reaction between the injected lime and CO_2. Since the calcium-based stage is not able to guarantee compliance with the IED emission limit for HCl (Fodor and Klemeš, 2012), HCl is further treated in a second, sodium-based stage, where sodium bicarbonate ($NaHCO_3$) is injected. As in the Ca-based stage, a fabric filter captures the process residues (mainly sodium chloride, NaCl). The Na-based residues can be sent to a dedicated recycling facility that regenerates fresh bicarbonate, while the residues from the Ca-based stage, which are mixed with fly ash from the combustion chamber, are unrecyclable and require disposal in proper sites (Margallo et al., 2015)

The analysed plant is subject to an annual mass flow limit for HCl emission equal to 1,000 kg/y. The HCl removal system operates at a constant set-point for HCl concentration at stack (position C3 in Figure 1) equal to 1 mg/Nm^3, while the set-point for HCl concentration downstream of the Ca-based stage (C2 in Figure 1) is set to 350 mg/Nm^3.
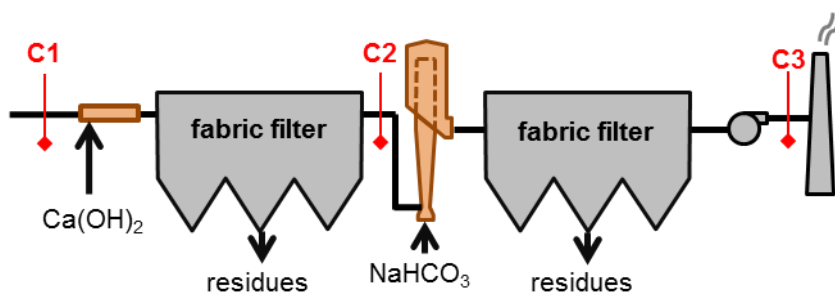

Figure 1: sketch of the two-stage HCl removal system. The three points C1, C2, C3 at which the concentration of HCl is measured are evidenced in red.

2.2 Process simulation

The following model, formulated in a previous study (Antonioni et al., 2012), was adopted in order to correlate the HCl removal efficiency X_{HCl} in a treatment stage with the required amount of reactant:

$$X_{HCl} = \frac{SR^n - SR}{SR^n - 1} \tag{1}$$

where the stoichiometric ratio *SR* is the ratio of the actual feed rate of reactant to the amount of reactant theoretically required to convert all the hydrogen chloride present in the flue gas and *n* is an empirical parameter, which needs plant-specific tuning. The parameter *n* was estimated for the Ca- and Na-based stages of the reference plant, on the basis of historical operational data. Details on the calibration of the model on the plant are reported elsewhere (Giannella, 2017), while the calibrated model is shown in Figure 2a. $Ca(OH)_2$ is evidently less reactive than $NaHCO_3$, due to the severe diffusional limitations that hinder its reactivity (Antonioni et al.,

2016). Furthermore, its reactivity towards HCl is heavily dependent on the operating temperature: lower temperatures are preferable, because a higher relative humidity has a promoting effect on the reaction (Chisholm and Rochelle, 1999).

The model was applied to simulate the operation of the two-stage system both for constant and variable set-points. A year of operation of the plant was considered and, for the sake of a simple presentation of the problem, the annual data were divided in 8 temporally consecutive periods, homogeneous in terms of HCl concentration entering the treatment system (C1) and operating temperature T of the Ca-based stage. The average values of HCl concentration at C1 and temperature are listed in Table 1. Figures 3a and 3b (shown in section 3) present the data averaged on a week basis, showing the seasonal variation of HCl concentration in the untreated flue gas and the fluctuation of T between maintenance events.

Table 1: Average values of HCl concentration at point C1 and operating temperature of the Ca-based stage for the 8 periods in which the year of operation was divided.

Period	1	2	3	4	5	6	7	8
C1 (mg/Nm^3)	1,115	867	880	622	782	793	856	1,205
T (°C)	191.4	182.0	192.9	182.4	188.1	178.9	178.3	186.0

In the case of constant set-points, given the HCl inlet concentration (C1) and having the set values of HCl concentration at C2 and C3 as mentioned in section 2.1, for each period the HCl removal efficiency required in the two stages is fixed.

For the cases with variable set-points, the following criteria were followed for the definition of the set-point:

- the variable set-point at stack (C3) was tuned to take into account the two fluctuating process conditions: T and HCl concentration at C1. For each period, given the yearly average μ for C1 (890 mg/Nm^3) and T (185.0 °C), if the period average for both variables is within a standard deviation σ of the yearly average the same set-point as the constant set-point case was adopted (1 mg/Nm^3). If at least one of the two variables is lower than μ – σ, a set-point of 1.5 mg/Nm^3 was chosen. Vice versa, if one of the two variables is higher than μ – σ, a set-point of 0.5 mg/Nm^3 was chosen. The chosen values respect the constraint of an identical annual mass flow emission of HCl for both a constant C3 and a variable C3 across periods.
- the variable set-point between the two stages (C2) was calculated according to a cost minimisation procedure. Given C1 and C3, the overall HCl removal efficiency is set, but there is a degree of freedom in the repartition of HCl removal between the stages, depending on the selected C2. For each period, C2 was chosen in order to minimise the operating costs of the line (see section 2.3 for cost entries). More details on the optimisation procedure are provided elsewhere (Dal Pozzo et al., 2016).
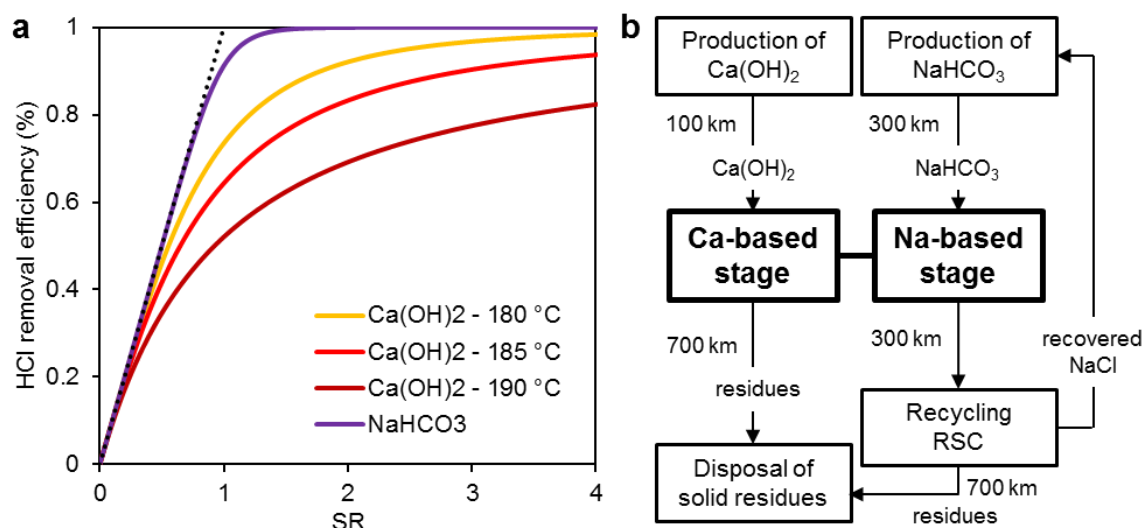

Figure 2: a) relationship between SR and HCl removal efficiency according to the calibrated model for the two reactants; b) system boundaries of the sustainability analysis.

2.3 Cost and impact assessment

The streams of reactants and residues required by the HCl removal system, quantified by the model, were used in order to estimate the associated costs and life cycle impacts. For the cost estimate, the following cost entries were considered: purchase costs for lime (50 €/t) and bicarbonate (240 €/t), disposal cost for the Ca-based residues (200 €/t) and recycling cost for Na-based residues (200 €/t).

Concerning the assessment of life cycle impacts, the system boundaries shown in Figure 2b were considered. Inventories for the unit processes included in the boundaries were retrieved from databases, like the European Life Cycle Database (ELCD, 2018).

Two categories of environmental impacts were taken into account: global warming potential (GWP) and acidification potential (AP). Characterisation factors for the two categories, respectively expressed in t CO_2 eq. and kg SO_2 eq., were retrieved from the widely adopted CML-IA database (CML, 2018).

3. Results

The results of the study are outlined in Figure 3, for the 8 periods in which the reference year was divided. Three cases were analysed: i) the operation with constant set-points at C2 and C3, which corresponds to the actual plant management; ii) the operation at constant HCl concentration at stack (C3) but variable concentration C2 between stages; iii) the operation at variable concentrations at both C2 and C3. The regulation of the set-points at C2 and C3 in the three cases is summarised in Figures 3c and 3d. The outcomes of the three cases are compared in terms of acidification potential (Figure 3e), global warming potential (Figure 3f) and operating costs (Figure 3g).

Firstly, it can be noted that, compared to the base case of constant C2 and C3 values, the operation with a variable set-point at C2 results in lower impacts and costs for every period. In this configuration, the possibility to reduce the utilisation of the Ca-based stage when the conditions are adverse to lime reactivity (*e.g.* in periods 1 and 3 with T > 190 °C) allows the system to avoid the significant over-stoichiometric feed rates of lime that are required in the base case. This translates in a lower global warming impact related to the carbon-intensive production of lime, which releases CO_2 during the calcination of limestone. Conversely, when the conditions are particularly favourable to lime reactivity (*e.g.* in period 6 with T < 180 °C and a low incoming HCl load), the configuration with variable C2 can increase the utilisation of the Ca-based stage, thus reducing the demand for bicarbonate and the acidification footprint associated with its chemical synthesis. Clearly enough, the optimised use of reactants generates lower operating costs.

When also the set-point for stack emission C3 is made variable, minor changes in the impact profiles can be observed. In particular, the global warming impact lowers when the set-point C3 is higher (less demand for reactants) and, vice versa, increases when the set-point C3 is lower. Conversely, the acidification impact increases when C3 increases and decreases when C3 decreases, because of the contribution of HCl emission to the impact category. The net result for both the global warming and the acidification categories when the numbers for each period are summed to give the overall outcomes for the entire year of operation (Table 2) is a slight reduction of the impacts compared to the case with variable C2 and constant C3.

More remarkably, Table 2 shows that it is possible to save 10 % of the HCl treatment cost and reduce respectively by 8 % and 13 % the life cycle acidification and global warming impacts of the two-stage treatment system, by introducing variable set-points. To put the numbers of Table 2 in perspective, it is worth noticing that the annual HCl mass flow of 1,000 kg/y the WtE plant can emit into atmosphere corresponds to 880 kg SO_2 eq., in terms of acidification potential. The reduction in terms of life cycle acidification impact obtained by introducing the variable set-points amounts to 397 kg SO_2 eq., which is equal to 45 % of the annual acidification impact generated by the emission of the WtE plant.

Therefore, the analysis arguably evidences the significant advantages of granting flexible operation to the treatment system. The work performed in this study presented only an 'ex-post' optimisation, in which the year of operation was divided in arbitrary periods for a clearer presentation of results. Based on the findings of this study, the implementation of a control strategy that continuously varies the set-points as a function of process variables such as temperature and inlet HCl load could be envisaged.

Table 2: Overall results for the entire year of operation of the reference plant in the three analysed cases.

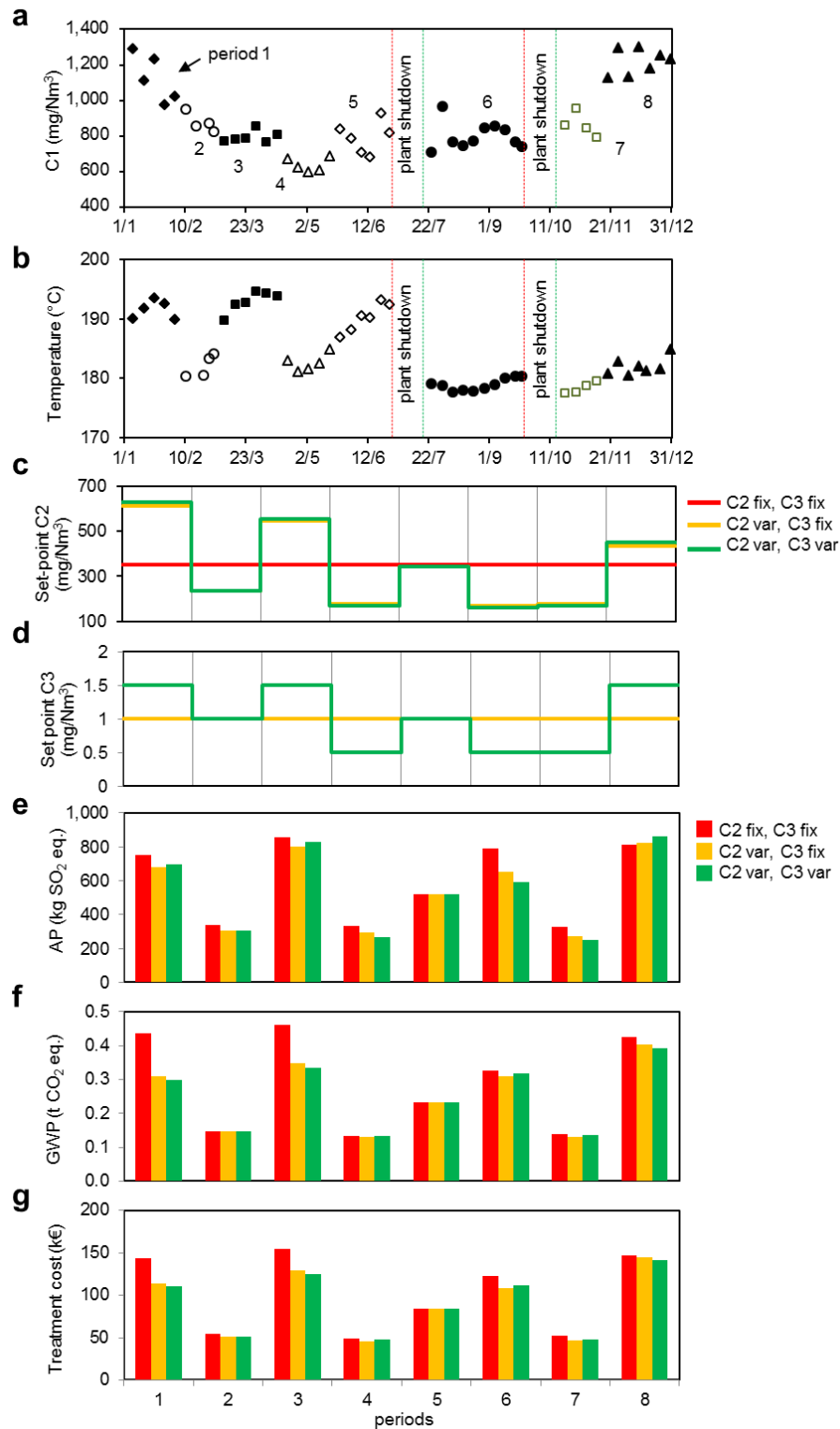| Cases	Constant C2 and C3	Variable C2, constant C3	Variable C2 and C3
AP (kg SO_2 eq.)	4,720	4,350	4,320
GWP (t CO_2 eq.)	2.30	2.01	1.99
Costs (k€)	806	722	717

Figure 3: a) inlet HCl concentration C1 (week average) for the reference year and division of the year in 8 periods; b) operating temperature of the Ca-based stage (week average) for the reference year; c) regulation of the set-point C2 in the 8 periods for the three cases; d) regulation of the set-point C3 in the 8 periods for the three cases; e) comparison of the total acidification potential in the period for the 8 periods for the three cases; f) comparison of the total global warming potential in the period for the 8 periods for the three cases; g) comparison of the total treatment costs in the period for the 8 periods for the three cases.

4. Conclusions

The present study was devoted to the assessment of the environmental and economic benefits of adopting variable set-points in the operation of a typical multi-stage HCl removal system.

The analysis evidenced a net reduction of environmental impacts and operating costs when variable set-points are introduced, because they allow an optimised use of the reactants. This approach is conscious of the variability of process conditions in the WtE environment and demands higher removal efficiencies when process conditions are favourable to HCl removal in order to counterbalance lower removal efficiencies when process conditions are adverse, thus maintaining the same annual target of HCl emission but with significantly lower indirect environmental burdens. This result can be safely generalised to WtE plants having two-stage dry HCl removal systems and mass flow emission limits equal or higher than the case study plant. Based on the observations of this study, the practical implementation at plant of control strategies considering variable set-points could allow to reduce overall environmental footprint of the process and improve its cost-effectiveness.

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