Optimization of the Economic and Environmental Profile of HCl Removal in a Municipal Solid Waste Incinerator through Historical Data Analysis

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In order to comply with increasingly ambitious environmental regulations, the best available techniques for the abatement of several airborne pollutants are often based on multi-stage treatment processes. Treatment of hydrogen chloride (HCl), a typical fuel-related pollutant from the combustion of waste or biomass, is such a case. In the multi-stage configuration, while the overall required removal of pollutants is fixed, there are degrees of freedom in the repartition of the removal between stages, leaving room for process optimization.

The present study offers an approach to the optimization of HCl removal, applied to a Municipal Solid Waste Incinerator (MSWI) equipped with a two-stage HCl abatement system based on the respective injection of hydrated lime and sodium bicarbonate. Historical plant data of the operation of the system for a reference year were collected and processed, in order to calibrate a semi-empirical model for the description of the two HCl treatment stages. The specific features of the two reactant systems, i.e. i) the temperature dependence of lime reactivity in the 1st stage and ii) the role of the bicarbonate cake on the fabric filter in the 2nd stage, were taken into account in the modelling.

Process operation was then simulated by means of the calibrated and validated model, thus showing the advantage in terms of cost savings (-15 % annual operating costs) and avoided environmental impacts (-21 % annual generation of process residues) of operating the system at its optimum compared to the recorded process performance. The optimal repartition of removal between stages was found to depend on operating temperature and inlet HCl concentration.

1. Introduction

Under the combined pressure of stricter national and local regulations and the increasing environmental awareness of the public opinion, the energy sector is increasingly adopting multi-stage treatment processes in order to ensure an enhanced removal efficiency of air pollutants. The abatement of acid gases like HCl, a signature pollutant from the combustion of waste (Hasan et al., 2017) or biomass, is typically performed by multi-stage dry or wet systems (Vehlow, 2015). These configurations ensure an outlet HCl concentration far below current legislative requirements (Directive 2010/75/EU), but they are often not operated at their optimum (De Greef et al., 2013). Besides guaranteeing emission compliance, a well-balanced utilization of the treatment stages should minimize the consumption of reactants and the generation of process residues, thus reducing both the associated costs and the indirect environmental burdens related to reactant production and residue disposal.

In the present study, an approach is proposed for the estimate of the optimal repartition of HCl removal between treatment stages. The reference case is a MSWI equipped with a two-stage lime/bicarbonate abatement system, which constitutes the state-of-the-art technology for dry acid gas removal (Dal Pozzo et al., 2016). The proposed approach is based on the collection of historical operational data for a reference year and their interpretation by means of a previously developed semi-empirical model, modified to take into account the specific reactivity features of the two reactants. The calibrated and validated model was thus applied to perform an 'ex post' optimization of the two-stage system in the reference year, in order to identify

the optimal operating point in terms of repartition of removal between 1st and 2nd treatment stages for different process conditions.

2. Modelling of HCl removal

2.1 Plant description

The analyzed plant is a medium-sized MSWI (56 MW$_{th}$) treating 16 t/h of waste and generating 130,000 Nm^3/h of flue gas. A two-stage dry treatment system is employed for HCl removal, as shown in Figure 1. The first treatment of the flue gas generated by waste combustion is performed by means of powdered hydrated lime, Ca(OH)$_2$. Downstream, a baghouse collects the process residues, known as residual calcium chemicals (RCC) and mainly formed by calcium hydroxychloride (CaOHCl), the solid product of the neutralization reaction between Ca(OH)$_2$ and HCl (Dal Pozzo et al., 2018a), some unreacted lime and the calcium carbonate (CaCO$_3$) generated by the undesired reaction between the injected lime and CO$_2$. The second treatment is based on the injection of sodium bicarbonate (NaHCO$_3$) in a dedicated reaction tower. In analogy with the 1st stage, a baghouse captures the process residues, known as residual sodium chemicals (RSC) and mainly formed by sodium chloride (NaCl).

The concentration of HCl, alongside the other relevant process variables, is monitored at three measurement points (C$_{HCl,1}$ at boiler exit, C$_{HCl,2}$ between treatment stages, and C$_{HCl,3}$ at stack). The plant is subject to a strict emission limit value in terms of HCl concentration at stack, equal to 0.5 mg/Nm^3 (20 times lower than the limit set by Directive 2010/75/EU).
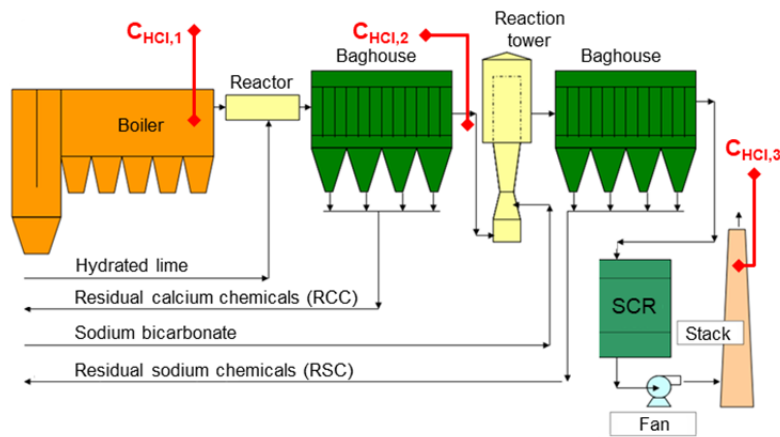

Figure 1: Outline of the considered flue gas cleaning line and measurement points of HCl concentration.

2.2 Model formulation and calibration

The relationship between the HCl removal efficiency X_{HCl} in a treatment stage and the required feed of reactant can be expressed by means of a semi-empirical model, developed in a previous work (Antonioni et al., 2012):

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

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 in the inlet flue gas and *n* is an empirical parameter, which needs plant-specific tuning.

In order to calibrate the model, process data of the treatment system (i.e. volumetric flow rate of the flue gas, HCl concentration and temperature at the three measurement points in Figure 1, as well as measured feed rates of Ca(OH)$_2$ and NaHCO$_3$) were collected on an hourly basis for a reference year. The >7500 hourly data points collected for each variable were divided in frequency classes, with the aim of excluding outliers and focusing the analysis on representative data of average process conditions.

For the calibration of the model for the 1st stage (hydrated lime), the 4 more frequent classes of inlet HCl concentration and operating temperature, highlighted in red in Figure 2, were considered. The wide range of variability of inlet HCl load is due to the variability of Cl content in the waste burnt (Dal Pozzo et al., 2017), while the operating temperature fluctuates as a consequence of fouling in the upstream heat recovery section

(Biganzoli et al., 2015). For each of the 16 groups of data resulting from the combination of the 4 classes of $C_{HCl,1}$ and the 4 classes of temperature, the averages of X_{HCl} and SR of lime feed were calculated. These 16 aggregated data points were used for model calibration, as shown in Figure 3a, where the model of Eq. (1) is presented on a linearized scale. It is evident from the aggregated process data that temperature plays a major role in the reaction between $Ca(OH)_2$ and HCl: at equal SR, a noticeably higher removal efficiency can be achieved when the operating temperature is lower. At lower temperatures, the relative humidity of the flue gas is higher and it is known from literature that moisture has a promoting effect on the reaction (Chisholm and Rochelle, 1999). Therefore, it resulted suitable to derive not a single calibrated value for the n parameter of the model, but 4 n values for the 4 classes of temperature. The resulting values were effectively approximated by the linear regression:

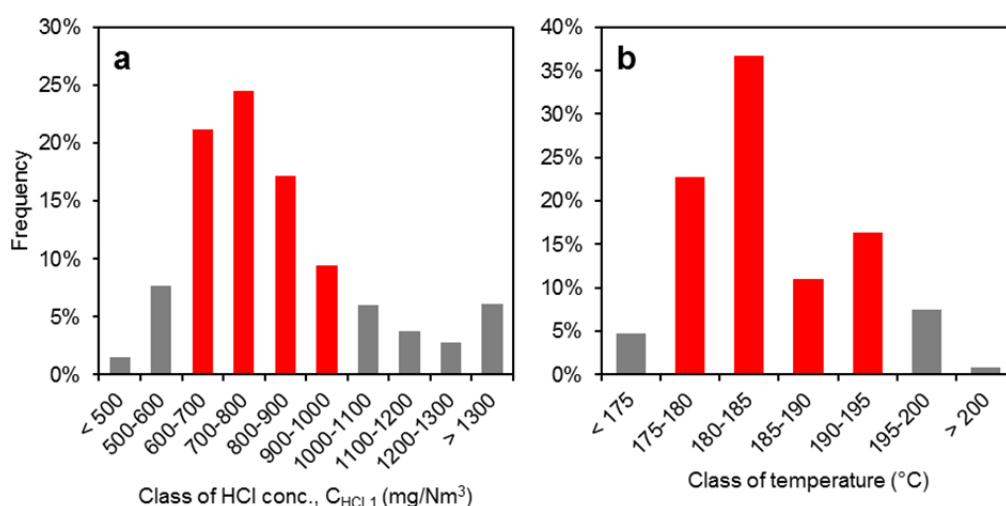$$n = 13.42 - 0.056 \cdot T(°C) \tag{2}$$

Figure 2: Distribution of a) inlet HCl concentration and b) operating temperature of the 1st treatment stage.

For the calibration of the model for the 2nd stage (sodium bicarbonate), a similar procedure was followed, by dividing the process data in 16 groups, calculating the 16 averages of X_{HCl} and SR, and using the resulting 16 data points for the calibration. As shown in Figure 3b, in this case a clustering of data as a function of temperature was not observed. Indeed, although the reactivity of $NaHCO_3$ can be slightly influenced by temperature, the temperature fluctuations in the 2nd stage are less pronounced (more than 80 % of data remains in the range 165-170 °C). Therefore, the entire dataset was used to extract a single parameter n (equal to 12.3) for the calibration of the model in Eq. (1). Nonetheless, a satisfactory fitting of data was not obtained. In particular, it appeared that, at low values of SR, the performance of the $NaHCO_3$-fed stage can exceed, for short times, the theoretical limit given by the complete consumption of the feed rate of reactant. This is arguably due to the fact that the overall HCl conversion is the sum of two contributions: i) the HCl neutralization performed by $NaHCO_3$ injected in the reaction tower, considered by the present model (conversion in reactor, X_R), and ii) a further neutralizing action performed by the cake of $NaHCO_3$ deposited over time on the bags of the baghouse downstream, which still retains a non-negligible sorption capacity (conversion in filter, X_F). Therefore, the model was reformulated in order to take into account both contributions:

$$X_{HCl} = X_R + X_F - X_R \cdot X_F = \frac{SR^n - SR}{SR^n - 1}(1 - X_F) + X_F \tag{3}$$

The model in Eq. (3), with the calibrated parameters n = 9.83 and X_F = 68 %, better describes the operational data (Figure 3b). For the $Ca(OH)_2$-fed stage, it was not necessary to introduce this model adaptation because, unlike for $NaHCO_3$, the solid conversion of lime is severely hindered by diffusional limitations (Antonioni et al., 2016) and the lime deposited on the baghouse is almost inert to further reaction. A further confirmation is given by the chemical composition of process residues. While RSC typically contains only the solid products of neutralization, RCC can retain a mass fraction of unreacted lime in the range 20-30 % (Bogush et al., 2015; Dal Pozzo et al., 2018b).
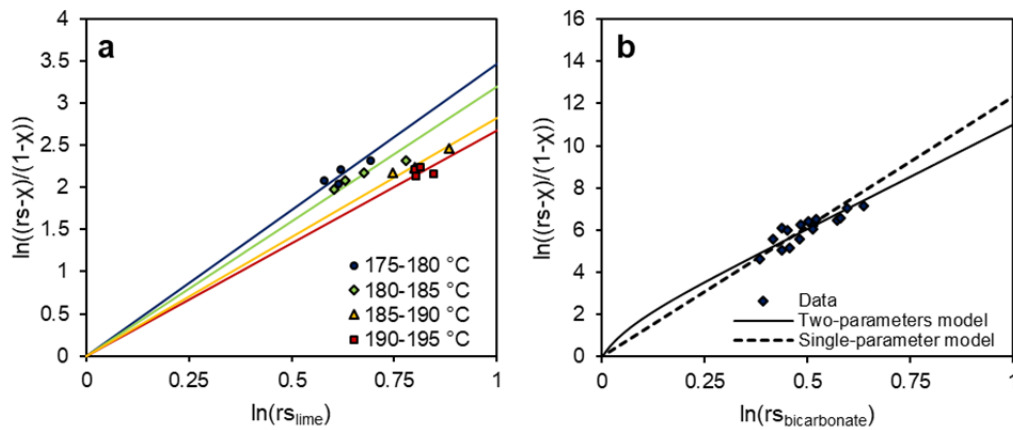
Figure 3: Model calibration for: a) 1st stage (hydrated lime); b) 2nd stage (sodium bicarbonate).

2.3 Model validation

The robustness of the calibrated model for the two treatment stages described in section 2.2 was tested against the process data of a reference year, grouped in eight successive time intervals (periods) characterized by homogeneous conditions in terms of inlet HCl concentration and operating temperature (Table 1).

Table 1: Partition of the reference year in the periods used for validation and optimization.

Period	Duration	Average $C_{HCl,1}$	T	Reduction of operating costs with optimization
	(dd/mm)	(mg/Nm^3)	(°C)	(%)
1	01/01-26/02	859	183	5.4
2	27/02-13/03	826	189	10.7
3	14/03-13/04	798	194	7.6
4	14/04-18/05	682	182	25.6
5	19/05-28/06	781	190	18.6
6	19/07-24/09	794	179	11.6
7	15/10-12/11	863	178	3.9
8	17/11-31/12	1097	182	17.0

The performance of the calibrated model in predicting the required feed rate of reactant, given the amount of HCl to remove and the operating temperature of the system, is shown by the parity plot in Figure 4, with reference to hydrated lime injection. The deviation between measured feed rates and predicted feed rates is in the range ±15 % for all the considered periods.
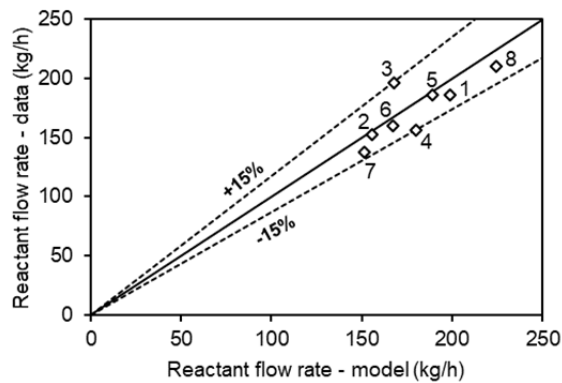

Figure 4: Model validation. Parity plot of the flow rate of hydrated lime: average process data in the periods of Table 1 vs. related model prediction.

3. Optimization of the process

Once tested the reliability of the model in reproducing plant operation, simulations were run to identify, for the same periods in which the reference year was divided for validation purposes (Table 1), what would have been the optimal repartition of HCl removal in the 1st and 2nd stage ('ex-post' optimization). For each period, the values of $C_{HCl,1}$ and T in Table 1 were considered, alongside the set value for $C_{HCL,3}$, and, while keeping constant the overall HCl removal efficiency of the line, the conversion of HCl in the 1st stage was varied in order to find, for each period, the degree of utilization of the 1st stage that minimises the operating costs.

The unit costs for the different cost entries were assumed as follows: 50 €/t for lime, 225 €/t for bicarbonate, 200 €/t for RCC disposal, 165 €/t for RSC recycling (RSCs can be sent to a dedicated recycling site which regenerates fresh bicarbonate; Dal Pozzo et al., 2017).

Figure 5 reports the results of the simulations for two periods, the 1st and the 4th in Table 1, by representing the variation of the overall operating costs as a function of HCl conversion in the 1st stage (at fixed overall HCl conversion in the two stages). In the plots, the diamond dot pinpoints the average operating point at which the plant operated during the period (historical data). The circle shows the optimal operating point. For all the eight periods, it was found that the minimization of costs is obtained for an utilization of the 1st stage lower than the one adopted in the plant. The related savings vary from period to period, as listed in Table 1, with a yearly average reduction of costs equal to 15 %. As detailed in panels b and d, the optimal operating point is characterized by a higher cost for NaHCO₃, due to the required higher utilization of the 2nd stage, which is more than compensated by marked savings in the disposal cost of RCCs, whose generation is significantly reduced. The economic optimization also entails an environmental optimization, since the generation of a non-recyclable hazardous waste such as RCC is one of the main environmental drawbacks of HCl dry treatment (Margallo et al., 2015): the yearly average reduction of process residues to disposal amounts to 21 %.

In order to generalize the results, the variation of the optimal HCl conversion in the 1st stage as a function of inlet HCl concentration ($C_{HCl,1}$) and operating temperature was studied more systematically generating the contour plot of Figure 6. A higher 1st stage is recommended when the temperature is lower (i.e., reactivity of lime is favoured) and when the inlet HCl load is higher (i.e., higher removal efficiency is required). In light of this result, it emerges clearly that a dynamic control of the repartition of HCl removal between the two stages as a function of process conditions is essential in achieving cost reduction.
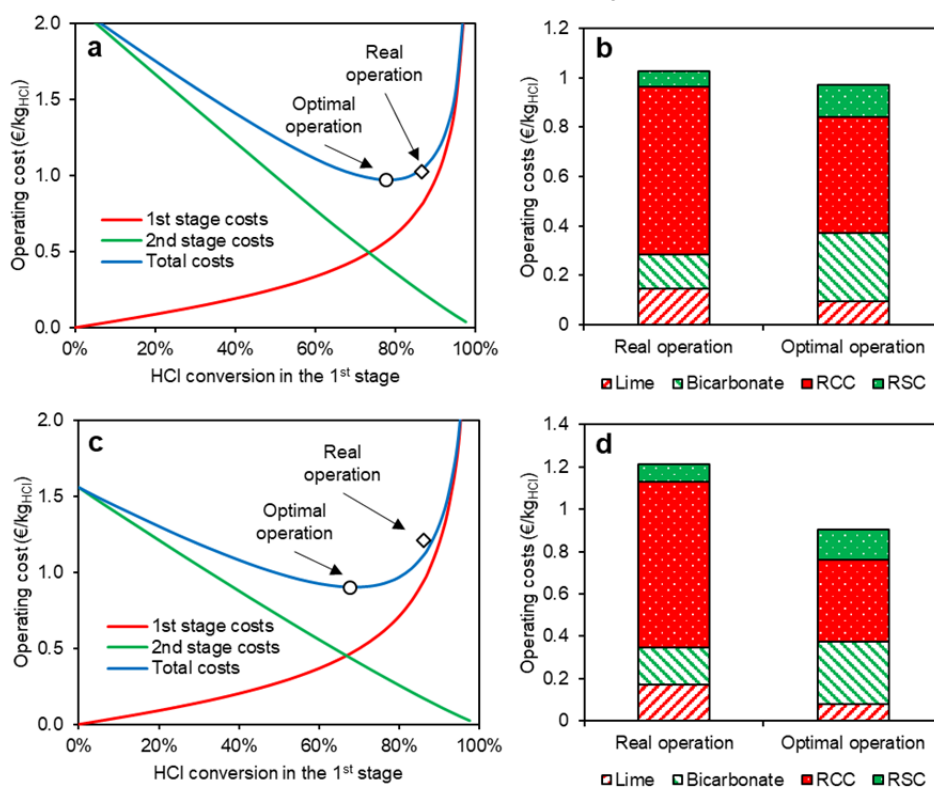

Figure 5: Optimization: variation of the operating costs as a function of HCl conversion in the 1st stage for period 1 (panel a) and period 4 (panel c) of Table 1. Comparison of the various cost entries for real and optimal operation in period 1 (panel b) and period 4 (panel d).
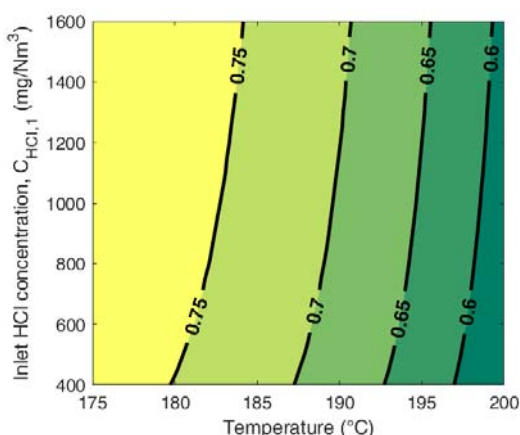
Figure 6: Optimal HCl conversion in the 1st stage as a function of inlet HCl concentration and operating temperature of the stage.

4. Conclusions

In the present work, a procedure based on the use of a semi-empirical model calibrated on historical process data, was used to identify the optimal operating configuration of a two-stage HCl removal system. The analysis evidenced the potential to save 15 % of operating costs as yearly average, if the system is operated at the optimal repartition of HCl conversion between the two stages. The savings are mainly due to the reduction of the process residues of hydrated lime injection, hence the solution that minimizes the costs realize also an environmental optimization by minimizing the amount of residues sent to disposal. The optimal operating point was found as a function of process temperature and HCl load entering the 1st treatment stage. Incorporating this function in the control system of the plant could allow to realize an 'ex ante' optimization, producing marked savings in the cost entries related to supply of reactants and disposal of residues.

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