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This is the final peer-reviewed author's accepted manuscript (postprint) of the following publication:

Published Version:

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 MANAGEMENT, 125, 303-315 [10.1016/j.wasman.2021.02.059].

Availability: This version is available at: https://hdl.handle.net/11585/821276 since: 2021-05-31

Published:

DOI: http://doi.org/10.1016/j.wasman.2021.02.059

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Economic and environmental benefits by improved process control strategies in HCl removal from waste-to-energy flue gas

HIGHLIGHTS

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- A methodology was outlined to test control strategies in operating facilities
- A virtual console was used to reproduce system behaviour
- Environmental, Economic and Technical performance indicators were defined
- Improved control strategies result in higher Environmental and Economic performance
- Full-scale test-runs confirmed the effectiveness of alternative control strategies

Economic and environmental benefits by improved process control strategies in HCl removal 1 2 from waste-to-energy flue gas 3 4 Alessandro Dal Pozzo, Giacomo Muratori, Giacomo Antonioni, Valerio Cozzani* 5 LISES - Dipartimento di Ingegneria Civile, Chimica, Ambientale e dei Materiali, Alma Mater 6 Studiorum - Università di Bologna, via Terracini n.28, 40131 Bologna, Italy 7 8 (*)corresponding author, Tel. +39-051-2090240, Fax +39-051-2090247, e-mail: valerio.cozzani@unibo.it 9

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11 Abstract

The control of HCl emission in waste-to-energy (WtE) facilities is a challenging flue gas 12 treatment problem: the release of HCl from waste combustion is highly variable in time and 13 14 the HCl emission standards are typically far lower in WtE than in any other industry. Traditional process control approaches in dry HCl removal processes are generally based on 15 16 feeding a large excess of solid reactants to the system, to ensure robustness and a wide safety margin in the compliance to environmental regulations. This results in the production of a 17 high amount of unreacted sorbents, strongly increasing the generation of solid wastes that 18 need to be disposed. In the present study, an approach was developed to allow the 19 20 implementation of improved control strategies for dry HCl abatement systems in operating full-scale facilities. Its objective is the reduction of the reactant feed and the waste 21 22 production, while still providing an adequate safety margin for emission compliance. The approach was based on the reproduction of the behaviour of the real system in a virtual 23 console that allows the extensive testing of alternative control strategies, limiting the need of 24 demanding test-runs at the real plant. A test case on an Italian WtE facility demonstrated the 25 capability of a control logic tuned in the virtual console to achieve a 13% reduction in the 26 consumption of reactants and generation of process residues, with unchanged HCl removal 27

efficiency. The results evidence the wide opportunities for optimisation of dry acid gas
removal systems, in particular when multistage systems are implemented.

30 **Keywords:** waste-to-energy, HCl, process optimization, dry sorbent injection.

31 **1** Introduction

In a modern waste management system, waste-to-energy (WtE) facilities have the role to 32 divert from landfilling waste streams for which recycling is currently technically or 33 34 economically unfeasible (Nizami et al., 2016) and enabling their thermal valorisation (Arena 35 et al., 2015), thus facilitating the transition to a circular economy (Bagheri et al., 2020; Van 36 Caneghem et al., 2019). Thanks to increasingly ambitious environmental regulations, the emission of several air pollutants related to WtE operation has been reduced more than 37 38 tenfold in the last decades (Ardolino et al., 2020; Damgaard et al., 2010). However, in the current holistic approach to environmental protection, the reduction of impacts has to go 39 40 beyond the minimisation of the emission of pollutants at the stack of the plant. Also indirect impacts, e.g. those associated to the consumption of reactants and the production of process 41 42 residues in the flue gas treatment system of the plant (Dal Pozzo et al., 2017; Dong et al., 2020; Lausselet et al., 2016), needs to be minimised. 43

Hydrogen chloride (HCl) is a typical pollutant in WtE flue gases, arising from the combustion
of waste containing chlorine (Zhang et al., 2019). Chlorine is widely dispersed amongst organic
and inorganic compounds present in several waste items (Gerassimidou et al., 2020; Yang et
al., 2018). Among the different techniques available for HCl removal (Bal et al., 2019; Dal
Pozzo et al., 2019; Ephraim et al., 2019; Kameda et al., 2020), dry sorbent injection (DSI) is
one of the technologies more frequently implemented (Beylot et al., 2018; Dal Pozzo et al.,
2018a). DSI consists in the in-duct addition of an alkaline powdered reactant (e.g. calcium)

hydroxide or sodium bicarbonate), which neutralises acid pollutants as HCl via gas-solid
reaction (Antonioni et al., 2016). DSI, adopted in either single or two-stage configurations (Dal
Pozzo et al., 2016; De Greef et al., 2013), is considered among the best available techniques
for flue gas treatment in WtE installations recommended by the European Union (Neuwahl et
al., 2019).

The main environmental drawback of DSI systems is the high stoichiometric excess of reactant feed that is required to achieve high HCl removal efficiency (Vehlow, 2015). The resulting excess consumption of reactant leads to the generation of relevant streams of solid process residues in the fabric filters, where they are collected together with fly ashes and micropollutants. The presence of these other components in the collected process residues causes the stream to be considered as hazardous waste and to require its disposal in dedicated landfill sites (Dal Pozzo et al., 2018b; Kameda et al., 2020).

63 In addition, given that the composition of the waste burnt in the combustion chamber of a 64 WtE plant varies widely over time, the resulting extreme variability of HCl concentration at the inlet of the flue gas treatment section (Dal Pozzo et al., 2020) is an inherent instability 65 66 that limits the effectiveness of conventional control strategies in calibrating the reactant feed needed to maintain a constant concentration setpoint at the outlet. Thus, the prevailing trend 67 in control strategies is to calibrate the process control parameters of the DSI system on the 68 69 safe side, and even more so accept high excess feed rates of reactants to minimise the 70 possible occurrence of overruns of HCl emission limits at stack.

A more accurate setting of the DSI control system could ensure not only a safe compliance of emission limits at stack, but also a reduction of the consumption of reactants and the generation of process residues. These in principle represent an undesired environmental

burden shift between different compartments (from air to soil/water) (Bogush et al., 2015;
Margallo et al., 2015; Quina et al., 2018).

The problem of the optimisation of flue gas treatment control with reference either to the 76 WtE context or to acid pollutants (HCl, SO₂, HF) is scarcely addressed in scholarly literature. 77 Ting et al. (2008) described the design of a PID control for acid gas removal via semi-dry 78 79 scrubbing in a WtE plant, with parameter tuning performed during commissioning operation. 80 Gassner et al. (2014) explored the use of data-driven modelling approaches to describe the 81 non-stationary operational behaviour of a semi-dry flue gas desulfurization process. Cignitti et al. (2016) developed a simple first principle model to predict the dynamics of a semidry SO₂ 82 83 absorber in desulfurization units of coal-fired power plants, while Guo et al. (2019) used a hybrid approach, blending first principles and neural network, to model and optimise a wet 84 flue gas desulfurization unit. Yet, the focus of these recent studies has been mainly the 85 86 theoretical development of enhanced dynamic models of the process, rather than their 87 implementation in real control schemes. In particular, to the best of the authors' knowledge, no previous paper addresses the potential environmental and economic advantages in terms 88 89 of reduced reactant consumption and related waste generation achievable with process control optimisation in WtE acid gas removal. 90

Furthermore, control optimisation in the WtE context is made complex by the fact that conventional direct tuning via extensive test runs during plant operation is generally incompatible with the need to comply with strict HCl emission limits in presence of a highly variable inlet load of HCl coming from waste combustion. In this regard, the set-up of datadriven simulations of the real system in a virtual environment, as more and more frequently performed in the manufacturing (Goodall et al., 2019) and process industry (Kockmann, 2019), could drastically reduce the need of field tests. By this strategy, the screening and the

tuning of new control settings is carried out directly in a virtual set-up, thus limiting the
number of in-field test runs only to those needed for the initial calibration of the simulation
and for the final trial of the new control system.

The present study focuses on the development of an approach for the optimisation of process 101 102 control in a typical DSI system for HCl removal based on a virtual environment. A dynamic 103 simulation of the dry treatment system was built in a virtual console implemented using the 104 Simulink software. A data-driven process model, calibrated with a specific set of test data, 105 nested into a reproduction of the control system of the DSI unit, was thus obtained and validated. The virtual console was used to test and tune an alternative control strategy, with 106 the objective to reduce the stoichiometric excess of reactant associated to HCl removal. The 107 108 alternative control was then tested in full scale at the real plant, demonstrating the potential 109 for significant environmental and economic benefits deriving from the reduction in reactant 110 consumption and related process waste generation.

112 2 Reference system and test facility

113

114 **2.1 HCl removal system**

The two-stage acid gas abatement system of a medium-sized (400 t/d of waste treated) WtE
facility located in Northern Italy was used as case study. As shown in Fig. 1, this system is
based on two consecutive steps of dry sorbent injection and filtration, taking place at ~180
°C, downstream of the heat recovery section of the plant. In the first stage, calcium hydroxide,
Ca(OH)₂, is injected, triggering the following gas-solid reaction of HCl neutralisation (lizuka et
al., 2020):

121
$$Ca(OH)_2 + 2 HCl \rightarrow CaCl_2 + 2 H_2O$$
 (1)

A fabric filter separates the solid product of reaction from the flue gas, together with a relevant unreacted fraction of Ca(OH)₂, present both due to the excess feed of reactant and for the intrinsic diffusional limitations of gas-solid reaction (i.e. the phenomenon of incomplete conversion discussed by Antonioni et al., 2016). In the second stage, the dry injection is based on sodium bicarbonate, NaHCO₃. At the injection temperature and, in general, at T > 130 °C (see Hartman et al., 2013), NaHCO₃ decomposes to porous sodium carbonate (Na₂CO₃), which in turn absorbs HCI (Dal Pozzo et al., 2019):

$$129 \quad 2 \, NaHCO_3 \, \rightarrow \, Na_2CO_3 \, + \, CO_2 \, + \, H_2O \tag{2}$$

130
$$Na_2CO_3 + 2 HCl \rightarrow 2 NaCl + CO_2 + H_2O$$
 (3)

Again, the solid product of reaction and an unreacted fraction of reactant are collected by a fabric filter. This two-stage configuration is adopted in several European WtE installations and it is appreciated for its built-in redundancy in terms of emission control (De Greef et al., 2013) and its flexibility that allows different repartitions of abatement demand between the two stages (Dal Pozzo et al., 2016). As shown in Fig. 1, the present study is focused on the optimisation of the control of the Ca(OH)₂ 1st stage of acid gas removal, referred to in the following as dry sorbent injection (DSI) system. As discussed in the following, the optimisation and tuning of the process control of the 1st stage not only improves the performance of the stage, but, stabilising the HCl outlet concentration, it also favours the optimal performance of the 2nd stage.

141

142 **2.2 Process control**

In the test facility, a conventional process control scheme implemented in several similar plants is present. The operation of the two-stage acid gas abatement system is monitored by the continuous acquisition of flue gas composition data at the measurement points PM1, PM2 and EM indicated in Fig. 1. The concentration of the main gas species at the sampling points, including the acid pollutants (HCl, SO₂, HF), is measured by Fourier-Transform infrared (FTIR) spectrometry, in compliance with CEN/TS 17337 (CEN, 2019), while the flue gas flowrate is determined at stack (point EM) by means of S-type Pitot tube velocity measurements.

150 In both the acid gas abatement stages, the distributed control system (DCS) of the plant 151 controls the solid reactant feed based on the measured inlet and outlet mass flowrates of 152 acid pollutants. A conditional logic selects the reactant feed rate as the maximum of two 153 values, calculated as follows:

Feedforward criterion. The calculated feed rate is equal to the stoichiometric demand
 related to the abatement of the inlet mass flowrates of acid pollutants at PM1,
 increased by a 10% excess.

ii. *Feedback criterion.* The feed rate is calculated according to a Proportional Integral (PI)
 feedback formula based on the difference between a set-point for the outlet HCI
 concentration and the value measured at PM2.

The settings of the feedback control (proportional gain K_P = 5 and integral gain τ_I = 8 s) 160 provide an aggressive reaction, i.e. strong excess feed rates of reactant are delivered 161 whenever the outlet HCl concentration exceeds the setpoint. Conversely, when the outlet HCl 162 concentration is lower than the setpoint, the feed rate of reactant does not drop as 163 significantly, because the feedforward criterion takes over. Thus, the combination of the 164 feedforward and feedback criteria as detailed above realises an asymmetrical control action, 165 166 in which the setpoint is actually treated as a threshold. The feedforward PI control works merely as an environmental safeguard, intended to act only if the feedforward is not capable 167 to maintain the outlet below the given threshold. A survey carried out by the authors involving 168 169 several Italian companies (HERAmbiente, HestAmbiente, IREN, Brianza Energia Ambiente) evidenced that this control strategy is typical of WtE acid gas abatement units, as the 170 objective is to avoid any spike in outlet HCl resulting from a variation in the inlet HCl load 171 172 coming from waste combustion (Muratori et al., 2020).

173

174 **2.3 Drawbacks of the reference control system**

175 The typical behaviour of the control system described in section 2.2 is shown in Fig. 2. Most of the time the control operates in feedforward mode and the feed rate of solid reactant is 176 proportional to the inlet HCl load. However, when the outlet HCl flowrate exceeds its setpoint, 177 178 the feedback mode takes over, imposing a relevant excess in feed rate to bring the HCl outlet 179 back under the threshold as soon as possible. This behaviour determines a peak in reactant consumption but generates also unintended instability in the outlet HCl flow rate. As 180 pinpointed by the arrows in Fig. 2, the spike of reactant feed manages to quickly reduce the 181 outlet HCl flow rate, but such a reduction is often followed by a swift rebound of outlet HCl 182 to high values that triggers another activation of the feedback control, resulting in another 183

spike of reactant feed. Since the layers of solid reactant accumulated over time on the fabric filter are known to play a major role in the overall acid gas removal action (Kim et al., 2017; Wu et al., 2004), the spikes of reactant feed might be detrimental because they induce unstable operation of the filter (Saleem and Krammer, 2012), activating frequent filter cleaning and reducing the residence time of reactant on the filter. The unstable HCl flow rate at the outlet of the 1st stage can in turn disturb the operation of the 2nd stage of acid gas removal.

In general, this control does not include the minimisation of reactant feed as a criterion anddoes not realise a rational use of reactant.

193

194 3 Methodology

195 **3.1 Framework**

196 Fig. 3 summarises the methodology developed to analyse the performance of alternative process control strategies for DSI, aimed at environmental and economic optimisation. The 197 198 core element of the methodology is the development of a process simulation that allows 199 exploring alternative control settings in a virtual console, while reducing the need for full-200 scale test-runs at the real plant. The process simulation duplicates into a software 201 environment the process units and the control system of the actual facility. As sketched in Fig. 3, building the simulation required: i) to reproduce the HCl removal process with a process 202 203 model; and ii) to simulate the control structure of the DSI unit. The first task required the 204 identification of an appropriate mathematical model for the description of the reaction 205 process (see section 3.2) and its training and validation on plant data collected from test-runs

(see section 3.3). The second task was performed replicating the control architecture of the
plant, briefly outlined in section 2.2, with a Simulink block diagram (see section 3.4).

The reliability of the simulation is validated considering the operating process control set-up in the real plant and comparing the outputs of the simulation with those recorded in the plant during normal operation, using the actual data as the input for the simulation. Once validated, the simulation can be used to screen and tune alternative control strategies, eventually leading to a new tuned control strategy that may be tested in the real plant, as in the test case that will be introduced in section 4.

Besides conventional indicators of process control performance, specific environmental and economic indicators (section 3.5) were defined to allow a comprehensive assessment of the performance of the alternative control strategies.

217

3.2 Selection of data-driven process model and input variables

219 As mentioned above, a mathematical model is required to reproduce the process dynamics 220 in the simulation. The process model needs to predict how the instantaneous HCl removal 221 efficiency varies depending on the inlet HCl concentration and the feed of solid reactant. Given the intrinsic unsteady nature of the process, this task can be addressed only with a 222 223 dynamic model capable of handling the rapidly changing operating conditions (e.g. variability 224 of HCl concentration due to variability of waste composition). Existing simplified stationary 225 models of acid gas removal that are typically applied for process optimisation studies (Harriott, 1990; Dal Pozzo et al., 2016) are clearly not apt for this task. On the other hand, 226 227 phenomenological models (Antonioni et al., 2016; Foo et al., 2017; Montagnaro et al., 2016) 228 that describe rigorously the kinetic and mass transfer phenomena involved in the gas-solid 229 reaction process were typically derived from laboratory-scale data and are not suitable to

simulate full-scale systems, as stated by Gutiérrez Ortiz and Ollero (2008) and Gassner et al.
(2014).

Therefore, a data-driven approach was chosen. A system identification procedure was 232 performed to estimate the structure and the parameters of the model from observed input-233 output plant data (Ljung, 2010). A simple input-output polynomial model, *i.e.* the linear auto-234 235 regressive exogenous (ARX) model, was selected as base for the system identification. ARX 236 models have already demonstrated to be reliable tools in emission control problems, e.g. in 237 the prediction of NO_x (Smrekar et al., 2013) or SO_x (Choi et al., 2002) emissions from coal-238 fired boilers. They are appreciated for their transparency and ease of interpretation (Akinola 239 et al., 2019). The general form of an ARX model is the following:

240

241
$$y(t) = a_1 y(t-1) + \dots + a_{n_a} y(t-n_a) + \sum_i \left[b_{1,i} u_i(t-n_k) + \dots + b_{n_{b,i}} u_i(t-n_{k,i}-n_{b,i}+1) \right] + e(t)$$
 (4)

242

where *y* is the output variable, u_i are the *i* input variables considered in the model, and *e* is the white-noise disturbance value. The values *a* and *b* are the model parameters, which can be represented in compact form in the parameter vector θ :

246

247
$$\theta = \begin{bmatrix} a_1 \cdots a_{n_a} \ b_{1,i} \cdots b_{n_{b,i}} \end{bmatrix}'$$
(5)

248

This model structure implies that the output variable *y* at time *t* is predicted as a linear combination of past output values (autoregressive part of the model) and current and past values of the input variables (exogenous part of the model). The parameters n_a and $n_{b,i}$ are, respectively, the number of past output samples and the number of past input samples (for each input variable *i*) considered for the prediction of the current output. The model can also consider input delay terms $n_{k,i}$, i.e. the number of input samples that occur before the input affects the output (also known as the dead time of the system). The use of past observations in the prediction of the output allows approximating also derivative terms by difference quotients, thus enabling the reproduction of the dynamics of the modelled system. The numbers n_a , $n_{b,i}$ and $n_{k,i}$ are known as hyperparameters and represent the order of the model, *i.e.* they indicate the number of parameters to optimise in the training of the model.

For the sake of simplicity, a two-input single-output ARX model was chosen for the present study. The modelled output *y* is the HCl molar flowrate in the flue gas leaving the DSI system. The two input variables u_i are the inlet HCl molar flowrate and the molar flowrate of Ca(OH)₂ fed to the DSI system.

In general, other variables might also affect the HCl removal process. The second most 264 abundant acid compound in WtE flue gases, SO₂, can consume a fraction of the reactant feed 265 266 (Zhang et al., 2019). Fluctuations in the flue gas flowrate can influence reactant residence 267 time (Hunt and Sewell, 2015). Variations in the operating temperature of the HCl removal stage, e.g. caused by fouling in the heat recovery section upstream, can alter the gas-solid 268 269 reaction kinetics (Dal Pozzo et al., 2018c). However, variations of temperature and flue gas flowrate are typically limited (see Fig. 2d and 2e) and, in the WtE plant under study, the inlet 270 SO2 concentration was a couple of orders of magnitude lower than that of HCl. Therefore, 271 272 these variables were excluded in the formulation of the model.

273

274 **3.3 Calibration of the model**

As a data-driven model, the ARX structure requires a specific calibration on data from the actual DSI system modelled. Informative data can be obtained by open-loop tests, in which

the control of the system is deactivated and process performance is assessed by varyingmanually the feed rate of reactant while recording inlet and outlet HCl concentration.

The dataset Z^N , formed by N consecutive observations of the input and output variables, obtained from the tests has to be divided in: i) a training set Z_{trn} , used for the estimation of the optimal model parameters; and, ii) a cross-validation set Z_{crv} , used for the selection of the optimal order of the model.

A further validation data set, *Z_{val}*, obtained collecting operating data from the normal, closedloop operation of the DSI system can be used for the assessment of the performance of the trained model.

286 Denoting as $\hat{y}(t|\theta)$ the output prediction of the model, least-square method is used to 287 estimate the parameter vector θ^* that produces the best fit of the training data Z_{trn} :

288
$$\theta^* = \arg\min\{V(\theta, Z_{trn})\}, \quad \text{where } V(\theta, Z_{trn}) = \frac{1}{N_{trn}} \sum_{t=0}^{N_{trn}-1} (y(t) - \hat{y}(t|\theta))^2 \quad (6)$$

The cross-validation compares the performance of models with different orders, each with its optimal parameter vector θ_i^* , estimated from the training set. The best model is the one for which $V(\theta, Z_{crv})$ is the smallest. This procedure helps selecting a model structure without unnecessary complexity (*i.e.* order), as excessively complex models tend to overfit the training set and perform poorly in the cross-validation set. Lastly, the model with order and parameters optimised for the Z_{trn} and Z_{crv} sets can be tested on the validation set Z_{val} and the procedure can go on iteratively until a given threshold of performance is fulfilled.

296

297 3.4 Virtual console

The process model described in section 3.2 was integrated into a simulation environment, where also the control loop and the other components of the DSI system were cloned as in

the real plant. The virtual console simulating the operation of the real DSI system consists offour blocks, as shown in Figure 4.

The block "*Data import*" defines the inlet conditions of the simulation (inlet HCl concentration and flue gas flowrate). These may be either actual plant data, collected at the measurement point PM1 (see Fig. 1), or artificial data, created to test the system performance under specific strain.

The input data of the "Data Import" block are then transferred to the "DTS" and "DCS" blocks. 306 307 The "DTS" block contains the process model described in section 3.2. The "DCS" block simulates the control system described in section 2.2. Specifically, given as input signals the 308 309 HCl load at the inlet of the DTS (provided by the "Data Import" block) and the HCl load at the outlet of the dry treatment system (modelled by the "DTS" block), this block evaluates with a 310 clock time of 1 s the command input for the actuator that regulates the feed rate of Ca(OH)₂. 311 312 The "Actuator" block simulates the operation of the screw feeder installed in the real plant. 313 The virtual actuator receives a percentage command of rotational speed calculated by the "DCS" block and transforms it into a molar feed rate of solid reactant to the "DTS" block, 314 315 following a linear relationship between percentage command and feed rate that is characteristic of the real feeder. The response of the actuator was modelled as a first order 316 317 transfer function:

318

319
$$G(s) = \frac{R}{T_m \cdot s + 1}$$
 (7)

320

321 where T_m is the actuation time of the screw feeder and R is the command to feed rate ratio.

This console allows the comparative testing of the behaviour of the DSI system under the default control (section 2.2) or an alternative control, as discussed in the test case described in section 4.

325

326 3.5 Performance indicators selected to test alternative control strategies

Both conventional indicators for process control performance and specific indicators capturing the environmental and economic performance of the process were defined to allow a comparison of alternative control strategies. The indicators are reported in Table 1 alongside their values obtained for the test case that will be introduced in section 4.

331 With respect to conventional process control indicators, these address the stability of the output variables. The instability of reactant injection, expressed as the ratio of the CV of 332 reactant injection to the CV of inlet HCl mass flow, measures the time variability of the feed 333 334 rate of reactant imposed by the control system. All things equal, a control demanding less 335 variable feed rates is preferred as it induces less mechanical stress on the feeding system. The instability of HCl outlet, expressed as the ratio of the CV of outlet HCl mass flow to the CV of 336 337 inlet HCl mass flow, measures the variability of the HCl mass flow at the outlet of the DSI system. 338

Environmental indicators trace the material streams responsible for the indirect environmental burdens of HCl removal: the *specific consumption of reactant*, expressed as mass of reactant injected per mass of removed HCl, and the *specific generation of residues*, expressed as mass of process residues generated per mass of removed HCl. These indicators were monitored both for the Ca-based 1st stage and the bicarbonate-fed 2nd stage of HCl removal, as the stabilisation of control in the 1st stage (object of the study) can also result in a more stable operation for the 2nd stage. Therefore, an indicator of *overall generation of*

residues, encompassing both treatment stages, was also considered to have a completepicture of the environmental benefit of control optimisation.

Lastly, an indicator addressing *overall operating costs* was also estimated, by translating the streams of reactants and residues in operating costs considering their unit costs (see Table S1).

351

352 4 Test Case

353

354 **4.1 Definition of the test case**

355 The test facility described in section 2.1 was used to define a test case for the application of the methodology outlined in section 3. An open-loop test-run was used for the calibration of 356 357 the ARX model, while the accuracy of the resulting virtual console in reproducing the system 358 behaviour under its default control was assessed using several datasets available for the 359 normal operation of the DSI system. An example of alternative control was proposed, tuned in the virtual console, then tested by full-scale test-runs on the real plant. The set of indicators 360 defined in section 3.4 was adopted to quantify the improvements in the stability of process 361 362 control and the economic and environmental performance.

363

4.2 Calibration of the model and validation of the simulation for the test case

The behaviour of the DSI system of the test facility was studied via step-response tests (Liu and Gao, 2012). Input excitations were applied to the system by varying stepwise the feed rate of Ca(OH)₂. The effect on system behaviour was recorded by continuous monitoring (30 s resolution time) of the outlet HCl concentration (measurement point PM2 in Fig. 1), while the inlet HCl concentration was also recorded (measurement point PM1 in Fig. 1). On the basis of the discussion provided in section 3.2, the ARX model was calibrated considering the molar flowrate of inlet HCl (calculated from the measured inlet HCl concentration and inlet flue gas flowrate) and the feed rate of Ca(OH)₂ as input variables, while the molar flowrate of outlet HCl (product of the measured outlet HCl concentration and outlet flue gas flowrate) is the modelled output.

The virtual console including the calibrated process model was then validated, comparing its simulated outlet of HCl with the measured values in four datasets of operation of the DSI system under the reference control, provided the same input data (see section 5.1). The simulation error was quantitatively assessed by calculating a cumulative normalised root mean squared error (RMSE):

380 Normalised RMSE (t) =
$$\frac{\sqrt{\frac{1}{n(t)}\sum_{i=1}^{n(t)}(y_i - \hat{y}_i)^2}}{\frac{\sum_{i=1}^{n(t)}y_i}{n(t)}}$$
(8)

381 where n(t) is the number of measurements/model evaluations at a given time.

382

383 **4.3 Selection and tuning of an alternative control**

384 Once the accuracy of the simulation results was demonstrated, the virtual console was used 385 to test alternative approaches to the control of HCl removal operation. In this test case, the control logic described in section 2.2 (named in the following as "conventional control") was 386 substituted with a simple feedback control (named in the following as "alternative control"). 387 388 Recalling Fig. 2, the conventional control is built to suppress any overrun of the setpoint of 389 outlet HCl concentration with a spike of Ca(OH)₂ feed. The consequences of such approach, 390 as illustrated in section 2.3, are an excess consumption of Ca(OH)₂ and unstable inlet conditions for the 2nd HCl removal stage fed with NaHCO₃, which, again, lead typically to an 391

excess consumption of NaHCO₃. Conversely, a properly tuned control in purely feedback
 action could limit the variability of both reactant feed and outlet HCl concentration.

The proposed feedback control is a simple proportional integral (PI) control. As the HCl inlet concentration signal is by nature highly variable and vulnerable to noise contamination (Coleman et al., 2019), the introduction of a derivative (D) control term was avoided, as it could generate system instability (Ting et al., 2008).

Hence, in the simulation the two parameters of the feedback control, K_P gain and τ_I integral

time, were tuned. The values of the optimised parameters were $K_P = 2$ and $\tau_I = 345$ s.

400

401 **4.4 Performance assessment of the new control at the real plant**

Eventually, a comparative assessment of the performance of the conventional and alternative control was carried out at the test facility. The alternative control was easily implemented, by deactivating the feedforward control and updating the feedback settings to the tuned parameters.

The test consisted in comparing a period of DSI process operation with the alternative control with a period of operation with the conventional control. The HCl load released by waste combustion can vary widely over time, and any control logic would manage better a low and uniform inlet mass flow of HCl, rather than a high and fluctuating one. Thus, to ensure a proper comparison, a period of operation experiencing an almost equal inlet mass flow of HCl to that present during the test of the alternative control was selected as representative of the conventional control performance.

As a measure of variability of inlet HCl load, the coefficient of variation (CV) of the HCl massflow during the test period was estimated:

415

416
$$CV = \frac{\sigma}{\mu}$$

where σ and μ are respectively the standard deviation and the mean of the measurements of inlet HCl mass flow during the period of study. It was also ensured that the two periods of DSI operation used for the comparison exhibited a similar CV of HCl mass flow, as it will be discussed in section 5.3. The HCl removal efficiency *X* was also calculated as follows:

$$422 X = \frac{m_{HCl,in} - m_{HCl,out}}{m_{HCl,in}} (10)$$

The comparison among the performance of the alternative control strategies was carried outcalculating the indicators discussed in section 3.5.

425

426 5 Results and Discussion

427 **5.1 Results of the validation of the simulation**

Figure 5 reports the performance of the virtual console in simulating the behaviour of the 428 conventional process control of the DSI system on a sample dataset (other samples are shown 429 430 in Figures S1-S3 in the Supporting Information, SI). The percentage command to reactant feed 431 given by the real system and by the simulation are compared in Fig. 5a. Figure 5b compares 432 the measured and the simulated outlet HCl mass flow. The yellow curve represents the set value of outlet HCl mass flow, which is a fluctuating value as it is defined as the product of the 433 434 fixed setpoint of outlet HCl concentration (see e.g. Fig. 2b) and the variable value of the flue gas flowrate (see e.g. Fig. 2d). Again, it can be noticed that the conventional control treats the 435 436 set value more like a threshold than a setpoint, as discussed in section 2.3. Figure 5c plots the cumulative average error of the simulation, represented as a normalised RMSE (introduced 437 in section 4.2). The error increases over time, indicating a loss of performance of the process 438

439 model nested in the simulation, that is typical of error accumulation in models of autoregressive nature (Bazghaleh et al., 2013; Nelles, 2020). As evidenced also by the figures 440 in the SI, the error grows faster when outlet HCl fluctuates widely, while it remains almost 441 442 unchanged and may even decrease during periods of stable operation. It is clear that a simple ARX model, linear by nature, falls short of achieving an accurate instantaneous prediction of 443 HCl outlet, which is the result of a complex and non-linear process involving gas-solid 444 445 reactions both in duct and on filter bags. Nonetheless, the simulation captures the average 446 system behaviour with acceptable resolution for the objective of the study.

447

448 **5.2** Results of the virtual testing of the alternative control

The simulation was used for the tuning and for the virtual testing of the alternative PI control. 449 The tuning of the alternative control by the methodology outlined in section 4.3 provided the 450 451 following value for the control parameters: proportional gain K_P = 2 and integral time τ_I of 452 345 s. It should be recalled that the PI settings of the feedback component of the conventional control (see section 2.2) are K_P = 5 and τ_I = 8 s. The alternative control is less aggressive, 453 454 with a reduced proportional action and a significantly higher integral time, which lowers the sensitivity of the control action to temporary deviations of the inlet HCl load. Figure 6a 455 illustrates the different behaviour of the alternative control strategy compared to the 456 457 conventional process control, on a data sample of 100 min. The simulation of the alternative 458 control was started during a significant deviation of the measured HCl outlet concentration from the set-point value to emphasise the different mode of operation of the two control 459 strategies. The feed rate variations imposed by the alternative control strategy are markedly 460 461 smoother than those of the conventional control. The proposed strategy accepts momentary 462 upticks in the HCl outlet concentration, whereas the action of the original control results in

spikes of reactant feed. Conversely, the alternative control strategy imposes a slightly higher feed rate than the original control during periods in which the latter operates in the feedforward mode. These opposite behaviours are evident from the plot of cumulated reactant consumption reported in Fig. 6c. Given that the variability of the reactant feed rate has been highlighted in section 2.3 as one of the main causes of inefficient reactant exploitation in the DTS, the alternative control strategy appears well suited to rationalise the use of the reactant, thus minimising the resulting generation of process residues.

470

471 **5.3 Results of the field test of the alternative control**

The alternative PI control was implemented in the DCS of the test facility. As outlined in section 4.4, a test run of the new control was carried out and the resulting operational data were compared with a previous period under the conventional process control configuration. The equivalence of action between the two controls was guaranteed by selecting the average value of outlet HCl concentration in the previous day under the conventional control as the setpoint for the test of the alternative control (see Fig. 7b).

478 Two 5-hour data samples with similar inlet flue gas conditions were selected for the comparative assessment. The two time series are shown in Fig. 7a, where it is possible to 479 compare qualitatively the behaviour of the two control strategies, i.e. the feed rate of 480 481 reactant and the outlet HCl flowrate, depending on the inlet HCl flowrate. The relative 482 performance of the two controls was tracked via the indicators introduced in section 3.5. Table 1 provides the list of the indicators used and the specific values obtained, while Figure 483 7c shows a radar plot comparing the normalised values of the performance indicators of the 484 485 alternative control to the reference one. Internal normalisation was used to obtain the values 486 shown in the figure. Given the low inlet SO₂ concentrations measured at the plant (in the range $10 - 30 \text{ mg/Nm}^3$) and the relatively low reactivity compared to HCl, the effect of SO₂ on system performance is negligible and not discussed in the analysis.

First of all, the two 5-hour data samples present highly comparable inlet HCl loads, hence the two controls are tested in a situation of similar stress. As reported in Table 1, the average inlet HCl mass flow rate in the two periods is equal and its CV is 68% higher during the test of the alternative control, i.e. the selection of data samples is slightly biased in favour of the conventional control.

Figure 7 shows that the real behaviour of the proposed PI-only control is in line with what was expected from the virtual simulation (see Fig. 6). The feed rate varies smoothly, with slow corrections in face of any sharp variation in the outlet HCl flow. Conversely, the conventional control reacts aggressively to deviations in the HCl outlet, with the characteristic spikes of reactant feed rate already described in Fig. 2.

When the performance indicators introduced in section 3.5 are considered, the parameter instability of reactant injection captures numerically this difference: while the commanded feed rate of the original control shows a CV that is 4.3 times higher than the CV of the inlet HCl molar flow, the CV of the commanded feed rate of the proposed control is only 1.24 times higher (a 71% reduction, see Table 1).

At the same time, the specific consumption of reactant in the Ca(OH)₂-fed treatment stage is 11% lower with the proposed control. This confirms that the lower aggressivity of the new control settings is not detrimental to the HCl removal efficiency of the system. On the contrary, in the test period, the proposed control managed to achieve the desired HCl removal performance with a significantly lower variability of reactant feed rate, which has the further advantage of reducing the mechanical stress to the screw feeder and the reactant transport system.

511 Another relevant metric is the instability of the outlet HCl flow, defined in section 3.5 as the ratio between the CVs of outlet and inlet HCl molar flow. The proposed PI-only control 512 achieves a 39% reduction in this indicator. This means that the HCl load exiting the Ca(OH)₂-513 514 fed treatment stage is less variable in time, thus the downstream NaHCO₃-fed stage operates on a less variable HCl inlet and is put in less stressful working conditions. As a consequence, 515 the optimisation of the control in the Ca(OH)₂ stage generates also a 26% reduction in the 516 517 specific consumption of reactant in the subsequent NaHCO₃ stage (see again Table 1), whose 518 control was not modified.

The overall consequence of the increase in efficiency owing to the new PI-only control is the 519 520 reduction in the production of the solid process residues of HCl removal via both the gas-solid reactions with $Ca(OH)_2$ and $NaHCO_3$. The new control achieves a 7% and a 22% reduction in 521 the generation of process residues, respectively in the 1st and 2nd treatment stages. The 522 523 overall effect is a 13% reduction of the amount of process waste generated by the HCl removal 524 operation. A further confirmation of this effect can be observed in figure S4 in the SI, which shows the simulated action of the conventional control system considering the inlet HCl load 525 526 for the 5-hour dataset collected during the test-run. The figure evidences that the multiple activations of the feedback mode would have caused a higher reactant consumption. 527

528

529 **5.4 Discussion**

530 In the light of the indicators in Table 1, the alternative control strategy tuned in the virtual 531 simulation was demonstrated to improve the overall economic and environmental 532 performance of the system. The consumption of reactants and the generation of process 533 residues were lowered in both the treatment stages, by increasing the efficiency of reactant 534 delivery. It was thus demonstrated that the main drawback of dry acid gas removal, i.e. the

required high excess of reactant, can be partially mitigated by introducing specific process control strategies. In particular, for a multistage system as that of the test facility, it is worth highlighting that an intervention limited to the 1st treatment stage can produce benefits also on the 2nd stage, by enabling a more efficient operation thanks to the lowered variability of the inlet HCI.

The alternative control strategy, based on a PI feedback control, however, has clear limitations: even if the simple feedback action reduces the variability of HCl load compared to the conventional control, the instability with respect to a setpoint is still quite high. More advanced control strategies could offer further improvements. Nonetheless, the proposed solution achieved the results in Table 1 with minimal need of full-scale testing and no significant change in the control architecture of the system, demonstrating the ease of implementation of better solid waste and reactant management via control optimisation.

The results obtained show that the procedure developed for the test of alternative control strategies, based on a virtual console, and the metric introduced, based on the performance indicators listed in Table 1, provide an effective approach to allow the improvement of the environmental and economic operational performance of acid gas treatment systems.

551

552 6 Conclusions

553 With increasingly strict limits on the emission of airborne pollutants as HCl, the flue gas 554 treatment sections in WtE installations are experiencing problems of excessive consumption 555 of reactants and related high generation of solid residues destined to landfilling, which lead 556 to non-negligible indirect environmental burdens. By considering a reference state-of-the-art 557 acid gas removal system, the present study demonstrated that a standard process control

approach based exclusively on the suppression of HCl emissions might be a suboptimal solution in terms of economic and environmental performance. A simple methodology based on virtual simulation and limited full-scale test-runs allowed identifying and tuning an alternative control strategy that achieved a reduction in the generation of solid process residues equal to 7% in the optimised Ca(OH)₂-fed 1st stage of HCl removal and 13% in the overall two-stage treatment line with respect to the original control configuration, while maintaining the same HCl emission performance at stack.

565 Despite the relevant advantages in terms of reactant economy, a limitation of the proposed solution is that it only partially alleviates the fluctuations in the HCl concentration at the outlet 566 of the 1st treatment stage, which are intrinsic to the WtE context. More advanced process 567 control strategies, taking into account process disturbances other than inlet pollutant 568 concentration and reactant feed rate, could be the key to develop plant-specific highly 569 570 performant model-based control schemes. However, the present study evidenced that 571 process control optimisation is a promising area of improvement in the management of WtE flue gas treatment, not only to improve stable operation, but also to increase significantly the 572 economic and environmental performance of DSI processes without hindering the 573 compliance to emission limits at stack. 574

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736 Figures and Tables





Figure 1. Scheme of the two-stage acid gas abatement system of the test facility considered,

740 including measurement points of flue gas composition (PM1, PM2 = process measurement,

741 EM = measurement at stack) and control loops for reactant feed rate. Control optimization of

742 1st stage (red box in the figure) was the object of the study.

743





Figure 2. Data recorded by the distributed control system (DCS) of an Italian WtE facility showing: a) the typical trend of inlet and outlet HCl flowrate and solid reactant feed rate during normal operation of the 1st stage acid gas removal unit applying the conventional process control strategy; b) threshold setpoint with respect to HCl inlet and outlet concentrations; c) SO₂ concentration; d) flue gas flowrate; e) operating temperature.



752 Figure 3. Methodology developed for testing and tuning of improved process control

753 strategies.

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Figure 4. Virtual console developed to simulate the DSI process (1st stage of the acid gas

removal system in Fig. 1) using the Simulink[®] software tool.



Figure 5. Performance of the virtual console in simulating the behaviour of the conventional
control of the system: a) measured vs. simulated command of reactant feed, b) measured vs.
simulated outlet HCl flow rate, compared to the set value of the control, c) cumulative average
error of the simulation.




Figure 6. a) Simulation of the reactant feed rate by the alternative PI control strategy
compared to the conventional process control on a sample dataset; b) deviation of the outlet
HCl concentration from the respective setpoints, c) resulting cumulated reactant consumption.



772

Figure 7. Results of test-run: a) 5-hour data samples obtained during alternative process control and conventional process control operation under a similar HCl input, b) outlet HCl concentration in the two 5-hour data samples, c) comparison between the normalised performance indicators of the alternative process control strategy to the conventional process control.

778	Table 1. Performance of the	alternative control strategy vs.	the conventional	process control

		Test period		
Parameter or indicator		Conventional control	Alternative control	Δ
Inlet HCI mass flow rate	μ	64.4	64.4	-
(kg/h)	CV	0.12	0.19	+68%
Feed rate of reactant, Ca(OH) ₂	μ	61.1	60.0	-2%
(kg/h)	CV	0.49	0.24	-52%
Outlet HCI mass flow rate	μ	17.2	12.5	-27%
(kg/h)	CV	0.51	0.52	+2%
HCl removal efficiency	μ	75.8	82.3	+9%
(%)	CV	11.2	8.5	-24%
Instability of reactant injection (CV of Ca(OH) ₂ feed rate / CV of inlet HCl)		4.27	1.24	-71%
Instability of HCl outlet (CV of outlet HCl / CV of inlet HCl)		4.4	2.7	-39%
Specific consumption of reactant (1 st : (kg of Ca(OH) ₂ fed / kg of HCl removed)	stage)	1.30	1.16	-11%
Specific generation of residues (1 st : (kg of residues / kg of HCl removed)	stage)	1.80	1.67	-7%
Specific consumption of reactant (2 nd : (kg of NaHCO ₃ fed / kg of HCl removed)	stage)	3.85	2.86	-26%
Specific generation of residues (2 nd) (kg of residues / kg of HCl removed)	stage)	2.55	2.00	-22%
Overall rate of residue production (kg of residues / kg of HCl removed)		2.00	1.73	-13%
Overall economic performance (€ of operating costs / kg of HCl removed)		1.86	1.48	-20%

779 monitored according to the performance indicators introduced in section 3.5.

Economic and environmental benefits by improved process control strategies in HCl removal 1 2 from waste-to-energy flue gas 3 4 Alessandro Dal Pozzo, Giacomo Muratori, Giacomo Antonioni, Valerio Cozzani* 5 LISES - Dipartimento di Ingegneria Civile, Chimica, Ambientale e dei Materiali, Alma Mater 6 Studiorum - Università di Bologna, via Terracini n.28, 40131 Bologna, Italy 7 8 (*)corresponding author, Tel. +39-051-2090240, Fax +39-051-2090247, e-mail: valerio.cozzani@unibo.it 9

10

11 Abstract

The control of HCl emission in waste-to-energy (WtE) facilities is a challenging flue gas 12 treatment problem: the release of HCl from waste combustion is highly variable in time and 13 14 the HCl emission standards are typically far lower in WtE than in any other industry. Traditional process control approaches in dry HCl removal processes are generally based on 15 16 feeding a large excess of solid reactants to the system, to ensure robustness and a wide safety margin in the compliance to environmental regulations. This results in the production of a 17 high amount of unreacted sorbents, strongly increasing the generation of solid wastes that 18 need to be disposed. In the present study, an approach was developed to allow the 19 20 implementation of improved control strategies for dry HCl abatement systems in operating full-scale facilities. Its objective is the reduction of the reactant feed and the waste 21 22 production, while still providing an adequate safety margin for emission compliance. The approach was based on the reproduction of the behaviour of the real system in a virtual 23 console that allows the extensive testing of alternative control strategies, limiting the need of 24 demanding test-runs at the real plant. A test case on an Italian WtE facility demonstrated the 25 capability of a control logic tuned in the virtual console to achieve a 13% reduction in the 26 consumption of reactants and generation of process residues, with unchanged HCl removal 27

efficiency. The results evidence the wide opportunities for optimisation of dry acid gas
removal systems, in particular when multistage systems are implemented.

30 Keywords: waste-to-energy, HCl, process optimization, dry sorbent injection.

31 1 Introduction

In a modern waste management system, waste-to-energy (WtE) facilities have the role to 32 divert from landfilling waste streams for which recycling is currently technically or 33 34 economically unfeasible (Nizami et al., 2016) and enabling their thermal valorisation (Arena et al., 2015), thus facilitating the transition to a circular economy (Bagheri et al., 2020; Van 35 36 Caneghem et al., 2019). Thanks to increasingly ambitious environmental regulations, the emission of several air pollutants related to WtE operation has been reduced more than 37 38 tenfold in the last decades (Ardolino et al., 2020; Damgaard et al., 2010). However, in the current holistic approach to environmental protection, the reduction of impacts has to go 39 beyond the minimisation of the emission of pollutants at the stack of the plant. Also indirect 40 impacts, e.g. those associated to the consumption of reactants and the production of process 41 42 residues in the flue gas treatment system of the plant (Dal Pozzo et al., 2017; Dong et al., 2020; Lausselet et al., 2016), needs to be minimised. 43

Hydrogen chloride (HCl) is a typical pollutant in WtE flue gases, arising from the combustion
of waste containing chlorine (Zhang et al., 2019). Chlorine is widely dispersed amongst organic
and inorganic compounds present in several waste items (Gerassimidou et al., 2020; Yang et
al., 2018). Among the different techniques available for HCl removal (Bal et al., 2019; Dal
Pozzo et al., 2019; Ephraim et al., 2019; Kameda et al., 2020), dry sorbent injection (DSI) is
one of the technologies more frequently implemented (Beylot et al., 2018; Dal Pozzo et al.,
2018a). DSI consists in the in-duct addition of an alkaline powdered reactant (e.g. calcium)

hydroxide or sodium bicarbonate), which neutralises acid pollutants as HCl via gas-solid
reaction (Antonioni et al., 2016). DSI, adopted in either single or two-stage configurations (Dal
Pozzo et al., 2016; De Greef et al., 2013), is considered among the best available techniques
for flue gas treatment in WtE installations recommended by the European Union (Neuwahl et
al., 2019).

The main environmental drawback of DSI systems is the high stoichiometric excess of reactant feed that is required to achieve high HCl removal efficiency (Vehlow, 2015). The resulting excess consumption of reactant leads to the generation of relevant streams of solid process residues in the fabric filters, where they are collected together with fly ashes and micropollutants. The presence of these other components in the collected process residues causes the stream to be considered as hazardous waste and to require its disposal in dedicated landfill sites (Dal Pozzo et al., 2018b; Kameda et al., 2020).

63 In addition, given that the composition of the waste burnt in the combustion chamber of a 64 WtE plant varies widely over time, the resulting extreme variability of HCl concentration at the inlet of the flue gas treatment section (Dal Pozzo et al., 2020) is an inherent instability 65 66 that limits the effectiveness of conventional control strategies in calibrating the reactant feed needed to maintain a constant concentration setpoint at the outlet. Thus, the prevailing trend 67 in control strategies is to calibrate the process control parameters of the DSI system on the 68 69 safe side, and even more so accept high excess feed rates of reactants to minimise the 70 possible occurrence of overruns of HCl emission limits at stack.

A more accurate setting of the DSI control system could ensure not only a safe compliance of emission limits at stack, but also a reduction of the consumption of reactants and the generation of process residues. These in principle represent an undesired environmental

burden shift between different compartments (from air to soil/water) (Bogush et al., 2015;
Margallo et al., 2015; Quina et al., 2018).

The problem of the optimisation of flue gas treatment control with reference either to the 76 WtE context or to acid pollutants (HCl, SO₂, HF) is scarcely addressed in scholarly literature. 77 Ting et al. (2008) described the design of a PID control for acid gas removal via semi-dry 78 79 scrubbing in a WtE plant, with parameter tuning performed during commissioning operation. 80 Gassner et al. (2014) explored the use of data-driven modelling approaches to describe the 81 non-stationary operational behaviour of a semi-dry flue gas desulfurization process. Cignitti et al. (2016) developed a simple first principle model to predict the dynamics of a semidry SO₂ 82 83 absorber in desulfurization units of coal-fired power plants, while Guo et al. (2019) used a hybrid approach, blending first principles and neural network, to model and optimise a wet 84 flue gas desulfurization unit. Yet, the focus of these recent studies has been mainly the 85 86 theoretical development of enhanced dynamic models of the process, rather than their 87 implementation in real control schemes. In particular, to the best of the authors' knowledge, no previous paper addresses the potential environmental and economic advantages in terms 88 89 of reduced reactant consumption and related waste generation achievable with process control optimisation in WtE acid gas removal. 90

Furthermore, control optimisation in the WtE context is made complex by the fact that conventional direct tuning via extensive test runs during plant operation is generally incompatible with the need to comply with strict HCl emission limits in presence of a highly variable inlet load of HCl coming from waste combustion. In this regard, the set-up of datadriven simulations of the real system in a virtual environment, as more and more frequently performed in the manufacturing (Goodall et al., 2019) and process industry (Kockmann, 2019), could drastically reduce the need of field tests. By this strategy, the screening and the

tuning of new control settings is carried out directly in a virtual set-up, thus limiting the
number of in-field test runs only to those needed for the initial calibration of the simulation
and for the final trial of the new control system.

The present study focuses on the development of an approach for the optimisation of process 101 102 control in a typical DSI system for HCl removal based on a virtual environment. A dynamic 103 simulation of the dry treatment system was built in a virtual console implemented using the 104 Simulink software. A data-driven process model, calibrated with a specific set of test data, 105 nested into a reproduction of the control system of the DSI unit, was thus obtained and validated. The virtual console was used to test and tune an alternative control strategy, with 106 the objective to reduce the stoichiometric excess of reactant associated to HCl removal. The 107 108 alternative control was then tested in full scale at the real plant, demonstrating the potential 109 for significant environmental and economic benefits deriving from the reduction in reactant 110 consumption and related process waste generation.

112 113

2 Reference system and test facility

114 **2.1 HCl removal system**

The two-stage acid gas abatement system of a medium-sized (400 t/d of waste treated) WtE
facility located in Northern Italy was used as case study. As shown in Fig. 1, this system is
based on two consecutive steps of dry sorbent injection and filtration, taking place at ~180
°C, downstream of the heat recovery section of the plant. In the first stage, calcium hydroxide,
Ca(OH)₂, is injected, triggering the following gas-solid reaction of HCl neutralisation (lizuka et
al., 2020):

121
$$Ca(OH)_2 + 2 HCl \rightarrow CaCl_2 + 2 H_2O$$
 (1)

A fabric filter separates the solid product of reaction from the flue gas, together with a relevant unreacted fraction of Ca(OH)₂, present both due to the excess feed of reactant and for the intrinsic diffusional limitations of gas-solid reaction (i.e. the phenomenon of incomplete conversion discussed by Antonioni et al., 2016). In the second stage, the dry injection is based on sodium bicarbonate, NaHCO₃. At the injection temperature and, in general, at T > 130 °C (see Hartman et al., 2013), NaHCO₃ decomposes to porous sodium carbonate (Na₂CO₃), which in turn absorbs HCI (Dal Pozzo et al., 2019):

$$129 \quad 2 \, NaHCO_3 \to Na_2CO_3 + CO_2 + H_2O \tag{2}$$

130
$$Na_2CO_3 + 2 HCl \rightarrow 2 NaCl + CO_2 + H_2O$$
 (3)

Again, the solid product of reaction and an unreacted fraction of reactant are collected by a fabric filter. This two-stage configuration is adopted in several European WtE installations and it is appreciated for its built-in redundancy in terms of emission control (De Greef et al., 2013) and its flexibility that allows different repartitions of abatement demand between the two stages (Dal Pozzo et al., 2016). As shown in Fig. 1, the present study is focused on the optimisation of the control of the Ca(OH)₂ 1st stage of acid gas removal, referred to in the following as dry sorbent injection (DSI) system. As discussed in the following, the optimisation and tuning of the process control of the 1st stage not only improves the performance of the stage, but, stabilising the HCl outlet concentration, it also favours the optimal performance of the 2nd stage.

141

142 **2.2 Process control**

In the test facility, a conventional process control scheme implemented in several similar 143 plants is present. The operation of the two-stage acid gas abatement system is monitored by 144 145 the continuous acquisition of flue gas composition data at the measurement points PM1, PM2 and EM indicated in Fig. 1. The concentration of the main gas species at the sampling points, 146 including the acid pollutants (HCl, SO₂, HF), is measured by Fourier-Transform infrared (FTIR) 147 148 spectrometry, in compliance with CEN/TS 17337 (CEN, 2019), while the flue gas flowrate is 149 determined at stack (point EM) by means of S-type Pitot tube velocity measurements. In both the acid gas abatement stages, the distributed control system (DCS) of the plant 150 151 controls the solid reactant feed based on the measured inlet and outlet mass flowrates of acid pollutants. A conditional logic selects the reactant feed rate as the maximum of two 152 values, calculated as follows: 153

Feedforward criterion. The calculated feed rate is equal to the stoichiometric demand
 related to the abatement of the inlet mass flowrates of acid pollutants at PM1,
 increased by a 10% excess.

ii. *Feedback criterion.* The feed rate is calculated according to a Proportional Integral (PI)
 feedback formula based on the difference between a set-point for the outlet HCI
 concentration and the value measured at PM2.

The settings of the feedback control (proportional gain K_P = 5 and integral gain τ_I = 8 s) 160 provide an aggressive reaction, i.e. strong excess feed rates of reactant are delivered 161 whenever the outlet HCl concentration exceeds the setpoint. Conversely, when the outlet HCl 162 concentration is lower than the setpoint, the feed rate of reactant does not drop as 163 significantly, because the feedforward criterion takes over. Thus, the combination of the 164 feedforward and feedback criteria as detailed above realises an asymmetrical control action, 165 166 in which the setpoint is actually treated as a threshold. The feedforward PI control works merely as an environmental safeguard, intended to act only if the feedforward is not capable 167 to maintain the outlet below the given threshold. A survey carried out by the authors involving 168 169 several Italian companies (HERAmbiente, HestAmbiente, IREN, Brianza Energia Ambiente) evidenced that this control strategy is typical of WtE acid gas abatement units, as the 170 objective is to avoid any spike in outlet HCl resulting from a variation in the inlet HCl load 171 172 coming from waste combustion (Muratori et al., 2020).

173

174 **2.3 Drawbacks of the reference control system**

175 The typical behaviour of the control system described in section 2.2 is shown in Fig. 2. Most of the time the control operates in feedforward mode and the feed rate of solid reactant is 176 proportional to the inlet HCl load. However, when the outlet HCl flowrate exceeds its setpoint, 177 178 the feedback mode takes over, imposing a relevant excess in feed rate to bring the HCl outlet 179 back under the threshold as soon as possible. This behaviour determines a peak in reactant consumption but generates also unintended instability in the outlet HCl flow rate. As 180 pinpointed by the arrows in Fig. 2, the spike of reactant feed manages to quickly reduce the 181 outlet HCl flow rate, but such a reduction is often followed by a swift rebound of outlet HCl 182 to high values that triggers another activation of the feedback control, resulting in another 183

spike of reactant feed. Since the layers of solid reactant accumulated over time on the fabric filter are known to play a major role in the overall acid gas removal action (Kim et al., 2017; Wu et al., 2004), the spikes of reactant feed might be detrimental because they induce unstable operation of the filter (Saleem and Krammer, 2012), activating frequent filter cleaning and reducing the residence time of reactant on the filter. The unstable HCl flow rate at the outlet of the 1st stage can in turn disturb the operation of the 2nd stage of acid gas removal.

In general, this control does not include the minimisation of reactant feed as a criterion anddoes not realise a rational use of reactant.

193

194 3 Methodology

195 **3.1 Framework**

196 Fig. 3 summarises the methodology developed to analyse the performance of alternative process control strategies for DSI, aimed at environmental and economic optimisation. The 197 198 core element of the methodology is the development of a process simulation that allows 199 exploring alternative control settings in a virtual console, while reducing the need for full-200 scale test-runs at the real plant. The process simulation duplicates into a software 201 environment the process units and the control system of the actual facility. As sketched in Fig. 3, building the simulation required: i) to reproduce the HCl removal process with a process 202 203 model; and ii) to simulate the control structure of the DSI unit. The first task required the identification of an appropriate mathematical model for the description of the reaction 204 205 process (see section 3.2) and its training and validation on plant data collected from test-runs

(see section 3.3). The second task was performed replicating the control architecture of theplant, briefly outlined in section 2.2, with a Simulink block diagram (see section 3.4).

The reliability of the simulation is validated considering the operating process control set-up in the real plant and comparing the outputs of the simulation with those recorded in the plant during normal operation, using the actual data as the input for the simulation. Once validated, the simulation can be used to screen and tune alternative control strategies, eventually leading to a new tuned control strategy that may be tested in the real plant, as in the test case that will be introduced in section 4.

Besides conventional indicators of process control performance, specific environmental and
economic indicators (section 3.5) were defined to allow a comprehensive assessment of the
performance of the alternative control strategies.

217

3.2 Selection of data-driven process model and input variables

219 As mentioned above, a mathematical model is required to reproduce the process dynamics 220 in the simulation. The process model needs to predict how the instantaneous HCl removal 221 efficiency varies depending on the inlet HCl concentration and the feed of solid reactant. Given the intrinsic unsteady nature of the process, this task can be addressed only with a 222 223 dynamic model capable of handling the rapidly changing operating conditions (e.g. variability 224 of HCl concentration due to variability of waste composition). Existing simplified stationary 225 models of acid gas removal that are typically applied for process optimisation studies (Harriott, 1990; Dal Pozzo et al., 2016) are clearly not apt for this task. On the other hand, 226 phenomenological models (Antonioni et al., 2016; Foo et al., 2017; Montagnaro et al., 2016) 227 228 that describe rigorously the kinetic and mass transfer phenomena involved in the gas-solid 229 reaction process were typically derived from laboratory-scale data and are not suitable to

simulate full-scale systems, as stated by Gutiérrez Ortiz and Ollero (2008) and Gassner et al.
(2014).

Therefore, a data-driven approach was chosen. A system identification procedure was 232 performed to estimate the structure and the parameters of the model from observed input-233 output plant data (Ljung, 2010). A simple input-output polynomial model, *i.e.* the linear auto-234 235 regressive exogenous (ARX) model, was selected as base for the system identification. ARX 236 models have already demonstrated to be reliable tools in emission control problems, e.g. in 237 the prediction of NO_x (Smrekar et al., 2013) or SO_x (Choi et al., 2002) emissions from coal-238 fired boilers. They are appreciated for their transparency and ease of interpretation (Akinola 239 et al., 2019). The general form of an ARX model is the following:

240

241
$$y(t) = a_1 y(t-1) + \dots + a_{n_a} y(t-n_a) + \sum_i \left[b_{1,i} u_i(t-n_k) + \dots + b_{n_{b,i}} u_i(t-n_{k,i}-n_{b,i}+1) \right] + e(t)$$
 (4)

242

where *y* is the output variable, u_i are the *i* input variables considered in the model, and *e* is the white-noise disturbance value. The values *a* and *b* are the model parameters, which can be represented in compact form in the parameter vector θ :

246

247
$$\theta = \begin{bmatrix} a_1 \cdots a_{n_a} \ b_{1,i} \cdots b_{n_{b,i}} \end{bmatrix}'$$
(5)

248

This model structure implies that the output variable *y* at time *t* is predicted as a linear combination of past output values (autoregressive part of the model) and current and past values of the input variables (exogenous part of the model). The parameters n_a and $n_{b,i}$ are, respectively, the number of past output samples and the number of past input samples (for each input variable *i*) considered for the prediction of the current output. The model can also consider input delay terms $n_{k,i}$, i.e. the number of input samples that occur before the input affects the output (also known as the dead time of the system). The use of past observations in the prediction of the output allows approximating also derivative terms by difference quotients, thus enabling the reproduction of the dynamics of the modelled system. The numbers n_a , $n_{b,i}$ and $n_{k,i}$ are known as hyperparameters and represent the order of the model, *i.e.* they indicate the number of parameters to optimise in the training of the model.

For the sake of simplicity, a two-input single-output ARX model was chosen for the present study. The modelled output *y* is the HCl molar flowrate in the flue gas leaving the DSI system. The two input variables u_i are the inlet HCl molar flowrate and the molar flowrate of Ca(OH)₂ fed to the DSI system.

In general, other variables might also affect the HCl removal process. The second most 264 abundant acid compound in WtE flue gases, SO₂, can consume a fraction of the reactant feed 265 266 (Zhang et al., 2019). Fluctuations in the flue gas flowrate can influence reactant residence 267 time (Hunt and Sewell, 2015). Variations in the operating temperature of the HCl removal stage, e.g. caused by fouling in the heat recovery section upstream, can alter the gas-solid 268 269 reaction kinetics (Dal Pozzo et al., 2018c). However, variations of temperature and flue gas flowrate are typically limited (see Fig. 2d and 2e) and, in the WtE plant under study, the inlet 270 SO2 concentration was a couple of orders of magnitude lower than that of HCl. Therefore, 271 these variables were excluded in the formulation of the model. 272

273

274 **3.3 Calibration of the model**

As a data-driven model, the ARX structure requires a specific calibration on data from the actual DSI system modelled. Informative data can be obtained by open-loop tests, in which

the control of the system is deactivated and process performance is assessed by varyingmanually the feed rate of reactant while recording inlet and outlet HCl concentration.

The dataset Z^N , formed by N consecutive observations of the input and output variables, obtained from the tests has to be divided in: i) a training set Z_{trn} , used for the estimation of the optimal model parameters; and, ii) a cross-validation set Z_{crv} , used for the selection of the optimal order of the model.

A further validation data set, *Z_{val}*, obtained collecting operating data from the normal, closedloop operation of the DSI system can be used for the assessment of the performance of the trained model.

286 Denoting as $\hat{y}(t|\theta)$ the output prediction of the model, least-square method is used to 287 estimate the parameter vector θ^* that produces the best fit of the training data Z_{trn} :

288
$$\theta^* = \arg\min\{V(\theta, Z_{trn})\}, \quad \text{where } V(\theta, Z_{trn}) = \frac{1}{N_{trn}} \sum_{t=0}^{N_{trn}-1} (y(t) - \hat{y}(t|\theta))^2 \quad (6)$$

The cross-validation compares the performance of models with different orders, each with its optimal parameter vector θ_i^* , estimated from the training set. The best model is the one for which $V(\theta, Z_{crv})$ is the smallest. This procedure helps selecting a model structure without unnecessary complexity (*i.e.* order), as excessively complex models tend to overfit the training set and perform poorly in the cross-validation set. Lastly, the model with order and parameters optimised for the Z_{trn} and Z_{crv} sets can be tested on the validation set Z_{val} and the procedure can go on iteratively until a given threshold of performance is fulfilled.

296

297 3.4 Virtual console

The process model described in section 3.2 was integrated into a simulation environment, where also the control loop and the other components of the DSI system were cloned as in

the real plant. The virtual console simulating the operation of the real DSI system consists offour blocks, as shown in Figure 4.

The block "*Data import*" defines the inlet conditions of the simulation (inlet HCl concentration and flue gas flowrate). These may be either actual plant data, collected at the measurement point PM1 (see Fig. 1), or artificial data, created to test the system performance under specific strain.

The input data of the "Data Import" block are then transferred to the "DTS" and "DCS" blocks. 306 The "DTS" block contains the process model described in section 3.2. The "DCS" block 307 simulates the control system described in section 2.2. Specifically, given as input signals the 308 309 HCl load at the inlet of the DTS (provided by the "Data Import" block) and the HCl load at the outlet of the dry treatment system (modelled by the "DTS" block), this block evaluates with a 310 clock time of 1 s the command input for the actuator that regulates the feed rate of Ca(OH)₂. 311 312 The "Actuator" block simulates the operation of the screw feeder installed in the real plant. 313 The virtual actuator receives a percentage command of rotational speed calculated by the "DCS" block and transforms it into a molar feed rate of solid reactant to the "DTS" block, 314 following a linear relationship between percentage command and feed rate that is 315 characteristic of the real feeder. The response of the actuator was modelled as a first order 316 317 transfer function:

318

319
$$G(s) = \frac{R}{T_m \cdot s + 1}$$
 (7)

320

321 where T_m is the actuation time of the screw feeder and R is the command to feed rate ratio.

This console allows the comparative testing of the behaviour of the DSI system under the default control (section 2.2) or an alternative control, as discussed in the test case described in section 4.

325

326 3.5 Performance indicators selected to test alternative control strategies

Both conventional indicators for process control performance and specific indicators capturing the environmental and economic performance of the process were defined to allow a comparison of alternative control strategies. The indicators are reported in Table 1 alongside their values obtained for the test case that will be introduced in section 4.

331 With respect to conventional process control indicators, these address the stability of the output variables. The instability of reactant injection, expressed as the ratio of the CV of 332 reactant injection to the CV of inlet HCl mass flow, measures the time variability of the feed 333 334 rate of reactant imposed by the control system. All things equal, a control demanding less 335 variable feed rates is preferred as it induces less mechanical stress on the feeding system. The instability of HCl outlet, expressed as the ratio of the CV of outlet HCl mass flow to the CV of 336 337 inlet HCl mass flow, measures the variability of the HCl mass flow at the outlet of the DSI system. 338

Environmental indicators trace the material streams responsible for the indirect environmental burdens of HCl removal: the *specific consumption of reactant*, expressed as mass of reactant injected per mass of removed HCl, and the *specific generation of residues*, expressed as mass of process residues generated per mass of removed HCl. These indicators were monitored both for the Ca-based 1st stage and the bicarbonate-fed 2nd stage of HCl removal, as the stabilisation of control in the 1st stage (object of the study) can also result in a more stable operation for the 2nd stage. Therefore, an indicator of *overall generation of*

residues, encompassing both treatment stages, was also considered to have a completepicture of the environmental benefit of control optimisation.

Lastly, an indicator addressing *overall operating costs* was also estimated, by translating the streams of reactants and residues in operating costs considering their unit costs (see Table S1).

351

352 4 Test Case

353

354 **4.1 Definition of the test case**

355 The test facility described in section 2.1 was used to define a test case for the application of the methodology outlined in section 3. An open-loop test-run was used for the calibration of 356 357 the ARX model, while the accuracy of the resulting virtual console in reproducing the system 358 behaviour under its default control was assessed using several datasets available for the 359 normal operation of the DSI system. An example of alternative control was proposed, tuned in the virtual console, then tested by full-scale test-runs on the real plant. The set of indicators 360 defined in section 3.4 was adopted to quantify the improvements in the stability of process 361 362 control and the economic and environmental performance.

363

4.2 Calibration of the model and validation of the simulation for the test case

The behaviour of the DSI system of the test facility was studied via step-response tests (Liu and Gao, 2012). Input excitations were applied to the system by varying stepwise the feed rate of Ca(OH)₂. The effect on system behaviour was recorded by continuous monitoring (30 s resolution time) of the outlet HCl concentration (measurement point PM2 in Fig. 1), while the inlet HCl concentration was also recorded (measurement point PM1 in Fig. 1).

On the basis of the discussion provided in section 3.2, the ARX model was calibrated considering the molar flowrate of inlet HCl (calculated from the measured inlet HCl concentration and inlet flue gas flowrate) and the feed rate of Ca(OH)₂ as input variables, while the molar flowrate of outlet HCl (product of the measured outlet HCl concentration and outlet flue gas flowrate) is the modelled output.

The virtual console including the calibrated process model was then validated, comparing its simulated outlet of HCl with the measured values in four datasets of operation of the DSI system under the reference control, provided the same input data (see section 5.1). The simulation error was quantitatively assessed by calculating a cumulative normalised root mean squared error (RMSE):

380 Normalised RMSE (t) =
$$\frac{\sqrt{\frac{1}{n(t)}\sum_{i=1}^{n(t)}(y_i - \hat{y}_i)^2}}{\frac{\sum_{i=1}^{n(t)}y_i}{n(t)}}$$
(8)

381 where n(t) is the number of measurements/model evaluations at a given time.

382

383 **4.3 Selection and tuning of an alternative control**

Once the accuracy of the simulation results was demonstrated, the virtual console was used 384 385 to test alternative approaches to the control of HCl removal operation. In this test case, the control logic described in section 2.2 (named in the following as "conventional control") was 386 substituted with a simple feedback control (named in the following as "alternative control"). 387 388 Recalling Fig. 2, the conventional control is built to suppress any overrun of the setpoint of 389 outlet HCl concentration with a spike of Ca(OH)₂ feed. The consequences of such approach, as illustrated in section 2.3, are an excess consumption of Ca(OH)₂ and unstable inlet 390 conditions for the 2nd HCl removal stage fed with NaHCO₃, which, again, lead typically to an 391

excess consumption of NaHCO₃. Conversely, a properly tuned control in purely feedback
action could limit the variability of both reactant feed and outlet HCl concentration.

The proposed feedback control is a simple proportional integral (PI) control. As the HCl inlet concentration signal is by nature highly variable and vulnerable to noise contamination (Coleman et al., 2019), the introduction of a derivative (D) control term was avoided, as it could generate system instability (Ting et al., 2008).

Hence, in the simulation the two parameters of the feedback control, K_P gain and τ_I integral

time, were tuned. The values of the optimised parameters were $K_P = 2$ and $\tau_I = 345$ s.

400

401 **4.4 Performance assessment of the new control at the real plant**

Eventually, a comparative assessment of the performance of the conventional and alternative control was carried out at the test facility. The alternative control was easily implemented, by deactivating the feedforward control and updating the feedback settings to the tuned parameters.

The test consisted in comparing a period of DSI process operation with the alternative control with a period of operation with the conventional control. The HCl load released by waste combustion can vary widely over time, and any control logic would manage better a low and uniform inlet mass flow of HCl, rather than a high and fluctuating one. Thus, to ensure a proper comparison, a period of operation experiencing an almost equal inlet mass flow of HCl to that present during the test of the alternative control was selected as representative of the conventional control performance.

As a measure of variability of inlet HCl load, the coefficient of variation (CV) of the HCl massflow during the test period was estimated:

415

416
$$CV = \frac{\sigma}{\mu}$$

417

418 where σ and μ are respectively the standard deviation and the mean of the measurements of 419 inlet HCl mass flow during the period of study. It was also ensured that the two periods of DSI 420 operation used for the comparison exhibited a similar CV of HCl mass flow, as it will be 421 discussed in section 5.3. The HCl removal efficiency X was also calculated as follows:

422
$$X = \frac{m_{HCl,in} - m_{HCl,out}}{m_{HCl,in}}$$
(10)

The comparison among the performance of the alternative control strategies was carried outcalculating the indicators discussed in section 3.5.

425

426 5 Results and Discussion

427 **5.1 Results of the validation of the simulation**

Figure 5 reports the performance of the virtual console in simulating the behaviour of the 428 conventional process control of the DSI system on a sample dataset (other samples are shown 429 430 in Figures S1-S3 in the Supporting Information, SI). The percentage command to reactant feed 431 given by the real system and by the simulation are compared in Fig. 5a. Figure 5b compares the measured and the simulated outlet HCl mass flow. The yellow curve represents the set 432 value of outlet HCl mass flow, which is a fluctuating value as it is defined as the product of the 433 fixed setpoint of outlet HCl concentration (see e.g. Fig. 2b) and the variable value of the flue 434 gas flowrate (see e.g. Fig. 2d). Again, it can be noticed that the conventional control treats the 435 436 set value more like a threshold than a setpoint, as discussed in section 2.3. Figure 5c plots the cumulative average error of the simulation, represented as a normalised RMSE (introduced 437 in section 4.2). The error increases over time, indicating a loss of performance of the process 438

model nested in the simulation, that is typical of error accumulation in models of 439 autoregressive nature (Bazghaleh et al., 2013; Nelles, 2020). As evidenced also by the figures 440 in the SI, the error grows faster when outlet HCl fluctuates widely, while it remains almost 441 unchanged and may even decrease during periods of stable operation. It is clear that a simple 442 ARX model, linear by nature, falls short of achieving an accurate instantaneous prediction of 443 HCl outlet, which is the result of a complex and non-linear process involving gas-solid 444 445 reactions both in duct and on filter bags. Nonetheless, the simulation captures the average 446 system behaviour with acceptable resolution for the objective of the study.

447

448 **5.2 Results of the virtual testing of the alternative control**

The simulation was used for the tuning and for the virtual testing of the alternative PI control. 449 The tuning of the alternative control by the methodology outlined in section 4.3 provided the 450 451 following value for the control parameters: proportional gain K_P = 2 and integral time τ_I of 452 345 s. It should be recalled that the PI settings of the feedback component of the conventional control (see section 2.2) are $K_P = 5$ and $\tau_I = 8$ s. The alternative control is less aggressive, 453 454 with a reduced proportional action and a significantly higher integral time, which lowers the sensitivity of the control action to temporary deviations of the inlet HCl load. Figure 6a 455 illustrates the different behaviour of the alternative control strategy compared to the 456 457 conventional process control, on a data sample of 100 min. The simulation of the alternative 458 control was started during a significant deviation of the measured HCl outlet concentration from the set-point value to emphasise the different mode of operation of the two control 459 strategies. The feed rate variations imposed by the alternative control strategy are markedly 460 461 smoother than those of the conventional control. The proposed strategy accepts momentary 462 upticks in the HCl outlet concentration, whereas the action of the original control results in

spikes of reactant feed. Conversely, the alternative control strategy imposes a slightly higher feed rate than the original control during periods in which the latter operates in the feedforward mode. These opposite behaviours are evident from the plot of cumulated reactant consumption reported in Fig. 6c. Given that the variability of the reactant feed rate has been highlighted in section 2.3 as one of the main causes of inefficient reactant exploitation in the DTS, the alternative control strategy appears well suited to rationalise the use of the reactant, thus minimising the resulting generation of process residues.

470

471 **5.3 Results of the field test of the alternative control**

The alternative PI control was implemented in the DCS of the test facility. As outlined in section 4.4, a test run of the new control was carried out and the resulting operational data were compared with a previous period under the conventional process control configuration. The equivalence of action between the two controls was guaranteed by selecting the average value of outlet HCl concentration in the previous day under the conventional control as the setpoint for the test of the alternative control (see Fig. 7b).

478 Two 5-hour data samples with similar inlet flue gas conditions were selected for the comparative assessment. The two time series are shown in Fig. 7a, where it is possible to 479 compare qualitatively the behaviour of the two control strategies, i.e. the feed rate of 480 481 reactant and the outlet HCl flowrate, depending on the inlet HCl flowrate. The relative performance of the two controls was tracked via the indicators introduced in section 3.5. 482 Table 1 provides the list of the indicators used and the specific values obtained, while Figure 483 7c shows a radar plot comparing the normalised values of the performance indicators of the 484 485 alternative control to the reference one. Internal normalisation was used to obtain the values 486 shown in the figure. Given the low inlet SO₂ concentrations measured at the plant (in the 487 range $10 - 30 \text{ mg/Nm}^3$) and the relatively low reactivity compared to HCl, the effect of SO₂ on
488 system performance is negligible and not discussed in the analysis.

First of all, the two 5-hour data samples present highly comparable inlet HCl loads, hence the two controls are tested in a situation of similar stress. As reported in Table 1, the average inlet HCl mass flow rate in the two periods is equal and its CV is 68% higher during the test of the alternative control, i.e. the selection of data samples is slightly biased in favour of the conventional control.

Figure 7 shows that the real behaviour of the proposed PI-only control is in line with what was expected from the virtual simulation (see Fig. 6). The feed rate varies smoothly, with slow corrections in face of any sharp variation in the outlet HCl flow. Conversely, the conventional control reacts aggressively to deviations in the HCl outlet, with the characteristic spikes of reactant feed rate already described in Fig. 2.

When the performance indicators introduced in section 3.5 are considered, the parameter instability of reactant injection captures numerically this difference: while the commanded feed rate of the original control shows a CV that is 4.3 times higher than the CV of the inlet HCl molar flow, the CV of the commanded feed rate of the proposed control is only 1.24 times higher (a 71% reduction, see Table 1).

At the same time, the specific consumption of reactant in the Ca(OH)₂-fed treatment stage is 11% lower with the proposed control. This confirms that the lower aggressivity of the new control settings is not detrimental to the HCl removal efficiency of the system. On the contrary, in the test period, the proposed control managed to achieve the desired HCl removal performance with a significantly lower variability of reactant feed rate, which has the further advantage of reducing the mechanical stress to the screw feeder and the reactant transport system.

511 Another relevant metric is the instability of the outlet HCl flow, defined in section 3.5 as the ratio between the CVs of outlet and inlet HCl molar flow. The proposed PI-only control 512 achieves a 39% reduction in this indicator. This means that the HCl load exiting the Ca(OH)₂-513 514 fed treatment stage is less variable in time, thus the downstream NaHCO₃-fed stage operates on a less variable HCl inlet and is put in less stressful working conditions. As a consequence, 515 the optimisation of the control in the Ca(OH)₂ stage generates also a 26% reduction in the 516 517 specific consumption of reactant in the subsequent NaHCO₃ stage (see again Table 1), whose 518 control was not modified.

The overall consequence of the increase in efficiency owing to the new PI-only control is the 519 520 reduction in the production of the solid process residues of HCl removal via both the gas-solid reactions with $Ca(OH)_2$ and $NaHCO_3$. The new control achieves a 7% and a 22% reduction in 521 the generation of process residues, respectively in the 1st and 2nd treatment stages. The 522 523 overall effect is a 13% reduction of the amount of process waste generated by the HCl removal 524 operation. A further confirmation of this effect can be observed in figure S4 in the SI, which shows the simulated action of the conventional control system considering the inlet HCl load 525 526 for the 5-hour dataset collected during the test-run. The figure evidences that the multiple activations of the feedback mode would have caused a higher reactant consumption. 527

528

529 **5.4 Discussion**

In the light of the indicators in Table 1, the alternative control strategy tuned in the virtual simulation was demonstrated to improve the overall economic and environmental performance of the system. The consumption of reactants and the generation of process residues were lowered in both the treatment stages, by increasing the efficiency of reactant delivery. It was thus demonstrated that the main drawback of dry acid gas removal, i.e. the

required high excess of reactant, can be partially mitigated by introducing specific process control strategies. In particular, for a multistage system as that of the test facility, it is worth highlighting that an intervention limited to the 1st treatment stage can produce benefits also on the 2nd stage, by enabling a more efficient operation thanks to the lowered variability of the inlet HCI.

The alternative control strategy, based on a PI feedback control, however, has clear limitations: even if the simple feedback action reduces the variability of HCl load compared to the conventional control, the instability with respect to a setpoint is still quite high. More advanced control strategies could offer further improvements. Nonetheless, the proposed solution achieved the results in Table 1 with minimal need of full-scale testing and no significant change in the control architecture of the system, demonstrating the ease of implementation of better solid waste and reactant management via control optimisation.

The results obtained show that the procedure developed for the test of alternative control strategies, based on a virtual console, and the metric introduced, based on the performance indicators listed in Table 1, provide an effective approach to allow the improvement of the environmental and economic operational performance of acid gas treatment systems.

551

552 6 Conclusions

553 With increasingly strict limits on the emission of airborne pollutants as HCl, the flue gas 554 treatment sections in WtE installations are experiencing problems of excessive consumption 555 of reactants and related high generation of solid residues destined to landfilling, which lead 556 to non-negligible indirect environmental burdens. By considering a reference state-of-the-art 557 acid gas removal system, the present study demonstrated that a standard process control

approach based exclusively on the suppression of HCl emissions might be a suboptimal solution in terms of economic and environmental performance. A simple methodology based on virtual simulation and limited full-scale test-runs allowed identifying and tuning an alternative control strategy that achieved a reduction in the generation of solid process residues equal to 7% in the optimised Ca(OH)₂-fed 1st stage of HCl removal and 13% in the overall two-stage treatment line with respect to the original control configuration, while maintaining the same HCl emission performance at stack.

565 Despite the relevant advantages in terms of reactant economy, a limitation of the proposed solution is that it only partially alleviates the fluctuations in the HCl concentration at the outlet 566 of the 1st treatment stage, which are intrinsic to the WtE context. More advanced process 567 control strategies, taking into account process disturbances other than inlet pollutant 568 concentration and reactant feed rate, could be the key to develop plant-specific highly 569 570 performant model-based control schemes. However, the present study evidenced that 571 process control optimisation is a promising area of improvement in the management of WtE flue gas treatment, not only to improve stable operation, but also to increase significantly the 572 economic and environmental performance of DSI processes without hindering the 573 compliance to emission limits at stack. 574

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736 Figures and Tables





739 **Figure 1.** Scheme of the two-stage acid gas abatement system of the test facility considered,

740 including measurement points of flue gas composition (PM1, PM2 = process measurement,

741 EM = measurement at stack) and control loops for reactant feed rate. Control optimization of

742 1st stage (red box in the figure) was the object of the study.

743





Figure 2. Data recorded by the distributed control system (DCS) of an Italian WtE facility showing: a) the typical trend of inlet and outlet HCl flowrate and solid reactant feed rate during normal operation of the 1st stage acid gas removal unit applying the conventional process control strategy; b) threshold setpoint with respect to HCl inlet and outlet concentrations; c) SO₂ concentration; d) flue gas flowrate; e) operating temperature.

750



752 Figure 3. Methodology developed for testing and tuning of improved process control

753 strategies.

754



756

Figure 4. Virtual console developed to simulate the DSI process (1st stage of the acid gas

removal system in Fig. 1) using the Simulink[®] software tool.


Figure 5. Performance of the virtual console in simulating the behaviour of the conventional
control of the system: a) measured vs. simulated command of reactant feed, b) measured vs.
simulated outlet HCl flow rate, compared to the set value of the control, c) cumulative average
error of the simulation.



Figure 6. a) Simulation of the reactant feed rate by the alternative PI control strategy
compared to the conventional process control on a sample dataset; b) deviation of the outlet
HCl concentration from the respective setpoints, c) resulting cumulated reactant consumption.



772

Figure 7. Results of test-run: a) 5-hour data samples obtained during alternative process control and conventional process control operation under a similar HCl input, b) outlet HCl concentration in the two 5-hour data samples, c) comparison between the normalised performance indicators of the alternative process control strategy to the conventional process control.

Parameter or indicator		Test period		_
		Conventional control	Alternative control	Δ
Inlet HCI mass flow rate (kg/h)	μ	64.4	64.4	-
	CV	0.12	0.19	+68%
Feed rate of reactant, Ca(OH)2 (kg/h)	μ	61.1	60.0	-2%
	CV	0.49	0.24	-52%
Outlet HCl mass flow rate (kg/h)	μ	17.2	12.5	-27%
	CV	0.51	0.52	+2%
HCl removal efficiency (%)	μ	75.8	82.3	+9%
	CV	11.2	8.5	-24%
Instability of reactant injection (CV of Ca(OH) ₂ feed rate / CV of inlet HCl)		4.27	1.24	-71%
Instability of HCl outlet (CV of outlet HCl / CV of inlet HCl)		4.4	2.7	-39%
Specific consumption of reactant (1 (kg of Ca(OH) ₂ fed / kg of HCl removed)	L st stage)	1.30	1.16	-11%
Specific generation of residues (1 (kg of residues / kg of HCl removed)	st stage)	1.80	1.67	-7%
Specific consumption of reactant (2 (kg of NaHCO ₃ fed / kg of HCl removed)	u nd stage)	3.85	2.86	-26%
Specific generation of residues (2 ¹ (kg of residues / kg of HCl removed)	nd stage)	2.55	2.00	-22%
Overall rate of residue production (kg of residues / kg of HCl removed)		2.00	1.73	-13%
Overall economic performance (€ of operating costs / kg of HCl remove	ed)	1.86	1.48	-20%

779 monitored according to the performance indicators introduced in section 3.5.

Declaration of interests

 \boxtimes The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

□The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Supplementary Material

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