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Wastewater management of wet scrubbers in waste-to-energy facilities: a life cycle analysis

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Nowadays, increasing attention is focused on the indirect impacts of emission control technologies and the potential burden shifting. A clear example is the generation of wastewater when wet scrubbing systems are used for flue gas cleaning. The present study takes into consideration a typical wet scrubber for waste-to-energy plants and the available options for wastewater management: physicochemical treatment, evaporation or valorisation. A comparative assessment of the environmental footprint of the alternatives is performed by taking into account the burdens related to the supply of reactants, energy and the disposal of process residues (solid and liquid waste streams). It is thus shown that the recovery of a marketable product from the effluent produces a net environmental benefit, while the choice between wastewater discharge or evaporation represents only a shift of burden between the water and the soil compartments.

1. Introduction

A key step of flue gas cleaning in waste incineration facilities is the removal of acid pollutants, mainly HCl and SO_2, released by the combustion of the Cl and S content in waste (Muratori et al., 2020; Zhang et al., 2019).
A wide array of techniques is available for the removal of acid gases, including the direct in-furnace injection of dolomitic sorbents (Dal Pozzo et al., 2020), the spray drying of lime slurries (Li et al., 2018), and the creation of a reactive filter cake at the baghouse filter via addition of dry powdered sorbents, such as calcium hydroxide (Dal Pozzo et al., 2018a; Tan et al., 2020), sodium bicarbonate (Dal Pozzo et al., 2019) or other compounds (Kameda et al., 2020; Wu et al., 2020).
Among these options, wet scrubbing is a widespread method of acid gas abatement, based on the physical or chemical absorption of acid pollutants in pure water or water with neutralizing additives (Vehlow, 2015). Although wet scrubbing systems are currently losing market shares to the aforementioned dry methods that eliminate water consumption (Gardoni et al., 2015) and exhibit lower operational costs (Dal Pozzo et al., 2018b), wet-based techniques are still widely appreciated for their unparalleled acid gas removal efficiency: they are often adopted in municipal solid waste incinerators and are the technology of choice for hazardous waste incinerators. The main drawback of wet scrubbing is the generation of an acid wastewater stream, also carrying trace contaminants such as heavy metals and organochlorinated compounds, that requires physicochemical treatment before safe discharge into public sewage systems.
In an integrated approach to pollution control, minimisation of any potential burden shift from an environmental compartment to another (here, from air to water) is paramount (Dal Pozzo et al., 2021). In that regard, different methods have been proposed to reduce the generation of wastewater of wet scrubbing systems. Although they all can bring significant benefits, a systematic evaluation of their full environmental impacts, which could help waste-to-energy (WtE) plant managers in the selection of the best suited technology for their specific situation, is lacking.
Such a variety of technological options can be categorised in two approaches. The elimination of a wastewater stream is pursued either by evaporation of the effluent or its transformation in a marketable product.
The present study selects relevant examples of these two approaches (and combination thereof) and performs a comparative assessment through a set of process and life cycle indicators to identify their environmental pros and cons.

2. Reference systems

2.1 Reference wet scrubber

A typical layout for an acid gas removal wet scrubbing system, outlined in Fig. 1, was chosen as reference scheme to provide a common basis for the comparative assessment of the alternatives for scrubber wastewater treatment.
Wet scrubbing is performed in two stages. In a first *acid scrubber* (a spray tower), hydrogen halides (mainly HCl, but also HF, HBr, HI in trace) are separated from the flue gas by physical absorption in water. For HCl:

$$HCl_{gas} \rightleftharpoons HCl_{solution} \rightleftharpoons H^+ + Cl^- \quad (1)$$

with the equilibrium almost entirely shifted to the product side (Vehlow, 2015).
In the second *neutral scrubber* (a packed bed vessel) the less water-soluble SO_2 is chemically absorbed by a solution of sodium hydroxide, NaOH:

$$SO_2 + 2\,NaOH \rightleftharpoons Na_2SO_3 + H_2O \quad (2)$$

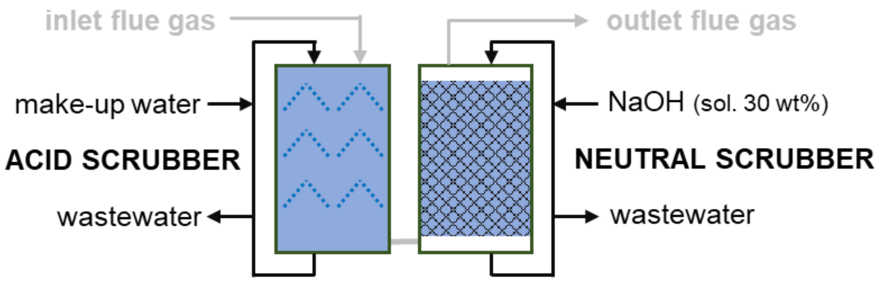

Figure 1: Reference scheme for the wet scrubbing system.

2.2 Alternatives for wastewater management

Four alternatives for wastewater management, as depicted in Fig. 2, are analysed in the present study.
The effluent of the acid scrubber typically presents a pH well below 1 (Vehlow, 2015), owing to HCl absorption, and contains small amounts of fine particulate matter and metal compounds. The effluent of the neutral scrubber contains mainly sodium sulphites/sulphates from reaction 2.
Conventionally, these effluents are mixed together and then sent to a pH correction stage, where $Ca(OH)_2$ is added to the wastewater to trigger the following reactions:

$$2\,HCl + Ca(OH)_2 \rightleftharpoons CaCl_2 + H_2O \quad (3)$$

$$Na_2SO_4 + 2\,HCl + Ca(OH)_2 \rightleftharpoons CaSO_4 + 2\,NaCl + \,2\,H_2O \quad (4)$$

The wastewater is then sent to a clariflocculator, where $CaSO_4$ is precipitated together with flocculated metal hydsetpoiroxides. The precipitated matter is dewatered and sent to hazardous landfill sites (Dal Pozzo et al., 2018c), while the treated wastewater is discharged in public sewers, provided that the chloride content is within acceptable limits. Fig. 2a summarises this common approach to wastewater management (Bc).
To avoid wastewater discharge, an alternative is to evaporate the effluent, typically by injection into the hot flue gas coming from the WtE boiler in a spray dryer (Ev, Fig. 2b). The dried residues are collected by a downstream fabric filter and then sent to hazardous landfill sites. This option still requires the pH correction tank but eliminates the clarifier and the wastewater discharge.
Recent attempts have been made to further minimise the amount of residues sent to disposal. Closing the chlorine cycle by recovering high-quality, marketable NaCl, HCl or Cl_2 from the drain of the acid scrubber is particularly challenging (Vehlow and Bourtsalas, 2017) and not addressed here. On the other hand, the valorisation of the effluent from the neutral scrubber is possible if H_2O_2 is used as absorbent instead of NaOH:

$$SO_2 + H_2O_2 \rightleftharpoons H_2SO_4 \quad (5)$$

The sulphuric acid at relatively low concentration generated by reaction 5 can be reused as such within the WtE plant (Andersson et al., 2014) or further concentrated to obtain a high-quality marketable product (see Sp, Fig. 2c). Lastly, evaporation of the effluent from the acid scrubber and valorisation of the effluent from the neutral scrubber can be combined in the solution depicted in Fig. 2d (Ev+Sp).

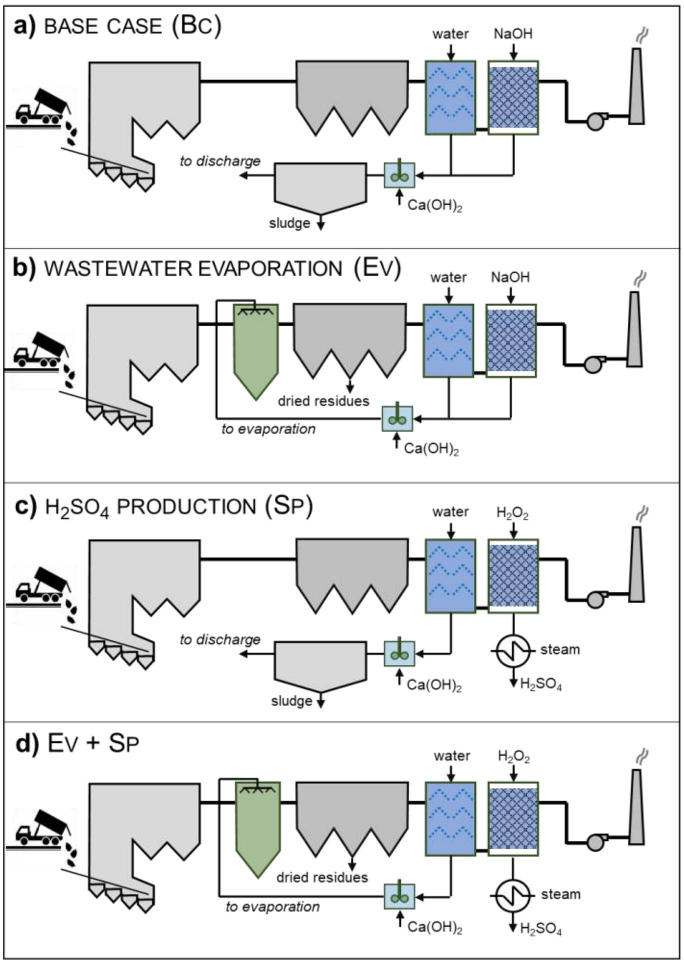

Figure 2: Schemes of the alternative of wastewater management considered in the study.

3. Modelling

3.1 Modelling of the wet scrubber

To conduct a quantitative assessment of the environmental consequences of the alternatives for wastewater treatment, the operation of the reference wet scrubber needs to be modelled. As reported in Table 1, two cases of flue gas composition, respectively representative of the typical acid gas load in flue gas from municipal solid waste combustion and from mixed industrial/urban waste combustion (Dal Pozzo et al., 2017; Romero et al., 2020), were studied. It was assumed that the wet scrubbing systems is installed in a medium-sized incinerator (100,000 Nm^3/h of flue gas) and the emission set-points for HCl and SO_2 are 0.5 and 2 mg/Nm^3, respectively.
The acid scrubber was modelled as an equilibrium stage at 60 °C. Considering the co-current flow of the flue gas stream G_s and the water stream L_s, the operation of the stage was described by the system:

$$Y_{eq} = f(X_{eq}) \tag{6}$$

$$G_s(Y_{eq} - Y_{in}) = L_s(X_{in} - X_{eq}) \tag{7}$$

where Y and X are respectively the HCl molar ratios in the gas and liquid phase. For the sake of simplicity, it was assumed that SO_2 does not undergo abatement in the acid stage. Eq. 6 followed Brandani et al. (1994).
It was imposed a pH set-point equal to 0.5 (Grieco and Poggio, 2009) in the recirculated water stream to calculate the flow rate of drained water. This value was then summed to an estimate of water evaporation in the scrubber to quantify the required flow rate of make-up water.
For the neutral scrubber, it was assumed that NaOH or H_2O_2 has to be fed with a 20% stoichiometric excess (Vehlow, 2015) to achieve the required SO_2 removal efficiency.
The power consumption of the scrubbing operation was estimated, considering two entries: the power required by the ID fan to overcome the pressure drop in the equipment (1.2 kPa, Neuwahl et al., 2019) and the power required for water pumping, considering a conservative estimate of 10 m for the suction head.

Table 1: Cases of inlet flue gas composition considered in the study.

Acid compound	Case 1	Case 2
HCl (mg/Nm^3)	600	1000
SO_2 (mg/Nm^3)	100	200

3.2 Modelling of the wastewater management options

For the conventional physicochemical treatment of scrubber wastewater (Bc), it was quantified the amount of hydrated lime required for the neutralisation of the pH of the effluent, according to reactions 3-4. Of the salts generated by pH correction, only $CaSO_4$ was assumed to precipitate and be collected as brine at the bottom of the subsequent clarifier. After dewatering of the brine in a filter press, it was assumed that the sludge retains a moisture content of 50% by weight (Metcalf & Eddy, Inc., 2003).
Conversely, wastewater evaporation by spray drying into hot flue gas (Ev) was assumed to obtain a dried solid residue with a moisture content < 5% by weight (Bianchini et al., 2015). The residues comprise both $CaSO_4$ and $CaCl_2$ from reactions 3-4. The thermal energy required to evaporate the effluent was accounted as penalty to heat recovery in the WtE plant.
In the case of H_2O_2 feed in the neutral scrubber and subsequent H_2SO_4 production (Sp), while the drain from the acid scrubber undergoes physicochemical treatment, the solution of H_2SO_4 from the neutral scrubber was assumed to require concentration up to 98.3% in order to produce a marketable product. The heat for water evaporation was assumed to be provided by a fraction of steam from the WtE boiler (Nilsson, 2020).
As for power consumption, it was estimated that the physicochemical treatment units require 3 MJ/m^3 wastewater (MetCalf & Eddy, Inc., 2003), while brine dewatering in filter press consumes 18 MJ/m^3 brine (Brinkmann et al., 2016).

3.3 Assessment of the environmental footprint

The comparative analysis of the environmental profile of the alternatives was performed by means of five indicators: water footprint (WF, expressed in m^3 of water consumed), carbon footprint (CF, in $kg_{CO2,eq}$) and cumulative energy demand (CED, in MJ of primary energy required) were used to assess the impacts arising from the life cycle of the material and energy streams needed for the operation of the wet scrubber and the wastewater management systems, while the quantification of the effluent discharge (in m^3) and the generation of solid residues (in kg) was used to evaluate the waste footprint of the wastewater management options.
Data on the WF, CF and CED of the reactants required for the analysed alternatives were elaborated starting from the life cycle inventories of the ecoinvent database (Frischknecht et al., 2005) and are summarised in Table 2. The values for H_2SO_4 are to be intended as avoided impacts of the industrial production of this compound, thanks to its generation as secondary product in the Sp and Ev+Sp options of Fig. 2.
The power and heat demand of the scrubber and the wastewater treatment options, calculated as outlined in section 3.1 and 3.2, was assumed to be supplied by the energy generation of the WtE plant, taking into account a typical 85% boiler efficiency (Viganò, 2018) and a 45% net electrical efficiency (Di Maria et al., 2016).

Table 2: Life cycle impacts associated to the production and supply of 1 kg of reactant.

Indicator		$Ca(OH)_2$	NaOH	H_2O_2	H_2SO_4
Carbon footprint	$kg_{CO2,eq}$	1.198	0.682	0.464	0.183
Water footprint	m^3 water	0.002	0.010	0.008	0.013
Cumulative energy demand	MJ	3.453	8.230	7.807	7.209

4. Results and discussion

Table 3 summarises the results from the mass and energy balances of the process alternatives following the approach of section 3. As outlined in section 2, the base case generates both a wastewater stream and a certain amount of solid residues. The main chemical consumed is $Ca(OH)_2$, given the relevant demand for pH neutralisation in the effluent, while the consumption of NaOH is comparatively limited, owing to the relatively low SO2 load in the flue gas and the high efficiency of reaction 2.
The Ev option eliminates the wastewater stream, at the cost of a small thermal energy penalty for the WtE plant and an increased generation of solid residues, owing to the precipitation of chlorides. Conversely, the Sp option retains the wastewater stream from the acid scrubber, but eliminates the residues from the neutral scrubber, at the cost of a thermal energy demand for H_2SO_4 concentration.

The process data of Table 3 were the basis for the calculation of the impact indicators introduced in section 3.3, which considered also the life cycle burdens related to the production of chemicals and energy. The indicators are shown in the radar plots of Fig. 3, normalised to the option with maximum value.

The base case exhibits high relative impacts in all categories, except for the generation of solid residues. Vice versa, the Ev option nullifies the impact for wastewater and markedly increases the generation of solid residues. From the point of view of the life cycle indicators (WF, CF and CED), the base case and the Ev case do not show significant differences.

Conversely, the Sp case is an all-round improvement compared to the base case, including in the life cycle indicators thanks to the avoided impacts associated with the production of marketable H_2SO_4. Likewise, the Ev+Sp case improves the results of the Ev case in all the categories.

Therefore, the analysis confirms that the modification of the neutral scrubber to produce H_2SO_4 generates a net environmental benefit, the additional impact related to energy penalty being more than offset by the avoided impacts of H_2SO_4 production. Instead, for the management of the effluent of the acid scrubber, there is no clear advantage for either wastewater discharge or evaporation. The preference between either choice is thus guided by site-specific considerations, e.g. on the vulnerability of local water systems or the availability of safe disposal sites for solid residues.

Table 3: Material and energy streams related to the alternatives under study (Fig. 2) for the two cases of flue gas composition in Table 1.

		Flue gas composition 1				Flue gas composition 2			
Item		Base case	Ev	Sp	Ev+Sp	Base case	Ev	Sp	Ev+Sp
Water	m^3/h	9.7	9.7	9.7	9.7	15.2	15.2	15.2	15.2
NaOH	kg/h	15.0	15.0	-	-	30.0	30.0	-	-
H_2O_2	kg/h	-	-	6.4	6.4	-	-	12.7	12.7
$Ca(OH)_2$	kg/h	77.6	77.6	66.0	66.0	138.8	138.8	115.6	115.6
Electricity	MJ/h	376.2	376.2	376.2	376.2	441.0	441.0	441.0	441.0
Heat	MJ/h	-	22.2	26.0	48.3	-	35.0	52.1	87.1
Wastewater	m^3/h	8.4	-	8.3	-	14.1		13.8	-
Residues [a]	kg/h	42.5	206.3	0.5	185.0	85.0	350.8	0.5	308.3
H_2SO_4 [b]	kg/h	-	-	15.3	15.3	-	-	30.6	30.6

[a] *sludge and/or solid residues from wastewater treatment*
[b] *sulphuric acid from wastewater valorisation*

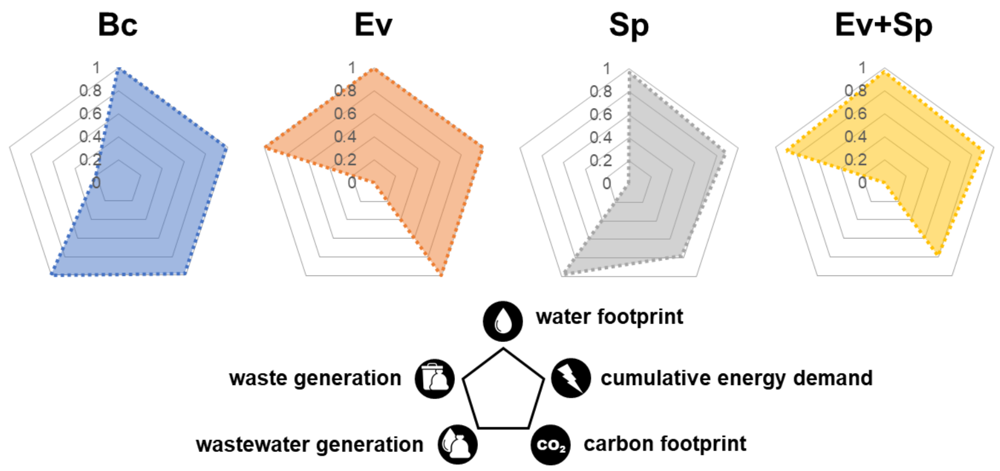

Figure 3: Normalised impact indicators for the alternatives under study. Case 1 of flue gas composition.

5. Conclusions

The present work took into consideration some relevant wastewater treatment options for the effluents of a typical WtE wet scrubbing system. The use of process and life cycle indicators allowed to trace a comprehensive profile of the environmental burdens associated to the analysed choices. The results suggest that the valorisation of the wastewater is a route that is advantageous on the environmental point of view. Conversely, the evaporation of wastewater realises a burden shift from wastewater to solid waste and its environmental advantage is not universal but might depend on local conditions.

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