

Alma Mater Studiorum Università di Bologna
Archivio istituzionale della ricerca

Assessment of cross-media effects deriving from the application of lower emission standards for acid pollutants in waste-to-energy plants

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

Published Version:

Dal Pozzo, A., Abagnato, S., Cozzani, V. (2023). Assessment of cross-media effects deriving from the application of lower emission standards for acid pollutants in waste-to-energy plants. *SCIENCE OF THE TOTAL ENVIRONMENT*, 856, 1-14 [10.1016/j.scitotenv.2022.159159].

Availability:

This version is available at: <https://hdl.handle.net/11585/899226> since: 2024-04-12

Published:

DOI: <http://doi.org/10.1016/j.scitotenv.2022.159159>

Terms of use:

Some rights reserved. The terms and conditions for the reuse of this version of the manuscript are specified in the publishing policy. For all terms of use and more information see the publisher's website.

This item was downloaded from IRIS Università di Bologna (<https://cris.unibo.it/>).
When citing, please refer to the published version.

(Article begins on next page)

Assessment of cross-media effects deriving from the application of lower emission standards for acid pollutants in waste-to-energy plants

Alessandro Dal Pozzo*, Samuele Abagnato, Valerio Cozzani

LISES - Dipartimento di Ingegneria Civile, Chimica, Ambientale e dei Materiali, Alma Mater Studiorum - Università di Bologna, via Terracini n.28, 40131 Bologna, Italy

(*) corresponding author, Tel. +39-051-2090296, e-mail: a.dalpozzo@unibo.it

Submitted for publication in *Science of the Total Environment*

Keywords: municipal solid waste; life cycle assessment; burden shifting; waste management; waste-to-energy.

Abstract

The recent release of the new European Commission reference document on the Best Available Techniques (BAT) for waste incineration has set ambitious targets for the control of the emission of pollutants. However, an improved performance of the existing flue gas treatment systems in waste-to-energy (WtE) facilities is usually associated to an increase of cross-media effects, i.e., additional indirect environmental impacts related to the increased consumption of reactants and to the increased generation of process residues/wastewater in flue gas treatment. The present study introduces an innovative approach to assess cross-media effects deriving from more stringent acid gas emission standards in the WtE sector. By coupling simplified process modelling and life cycle analysis, the proposed methodology links the higher removal efficiency required for flue gas treatment to the impacts related to the reactants supply and waste disposal chain. An application to the Italian WtE sector exemplifies the potential of the method. The results evidence that, in case of HCl emission setpoints lower than 1 mg/Nm³, the reduction of acidifying emissions at the WtE stacks can be offset by the increase of global warming and smog formation impacts in the supply chain of

flue gas cleaning reactants. In case of setpoints lower than 0.5 mg/Nm^3 , even within the acidification category the increase of indirect impacts more than compensates the decrease of WtE emissions. The net environmental benefit is strongly affected by the type of acid gas removal technology adopted, with dry systems typically associated with a larger increase of cross-media burdens when required to perform at higher removal efficiencies.

1. Introduction

In 2018, 28% of the 250 million tons of municipal solid waste (MSW) generated in the European Union (EU) were treated in waste-to-energy (WtE) plants (EUROSTAT, 2019). EU member states waste management policy aims at progressively phasing out landfilling (Ferreira et al., 2017; Wang et al., 2020) and avoiding the dispersion of waste streams to the environment (Horton et al., 2017). Thus, MSW incineration will still play a key role in the future, ensuring that all the residual waste that cannot be recycled is safely processed, harnessing its residual value as energy for district heating and electricity generation (Ali et al., 2020; Van Caneghem et al., 2019).

However, a primary requirement for WtE sustainability is that appropriate flue gas treatments minimize the emission of air pollutants generated in MSW combustion (Malinauskaite and Jouhara, 2019). In order to minimize the environmental impact of industrial activities, the European Union has issued sectorial documents identifying the Best Available Techniques (BAT) for pollution control, often associated to emission levels (BAT-AELs). The BAT-AELs are mandatory guidelines that local authorities use to define the specific emission limit values (ELVs) of new or operating facilities. The progressive updating of BAT documents, due to the advancements in process technologies, results in a progressive reduction of BAT-AELs.

Among others, acid gases (mainly HCl and SO₂) are a class of macro-pollutants generated in waste incineration, stemming from the combustion of waste fractions containing chlorine and sulphur (Gerassimidou et al., 2021, Zhang et al., 2019). HCl and SO₂ are precursors of environmental acidification (Evans et al., 2011). **Table 1** reporting the recently revised BAT-AELs for acid gases emissions in WtE operation implemented by the EU (2019 BREF WI; European Commission et al., 2020) and the previous BAT-AEL values provides a clear example of the progressive reduction policy of BAT-AELs. The new BAT-AELs are thus the reference value for the ELVs imposed to acid gases emissions in new or existing WtEs (in the latter case, by the renewal process of operating permits, that should be concluded by 2024). Thus, by 2024 the implementation of the 2019 BREF WI is expected to result in a significant reduction of the environmental impact due to acid gas emissions by WtE facilities.

Table 1. Previous and current BAT-AELs related to HCl and SO₂ for waste incineration plants, expressed as daily average values.

Parameter	Unit	Previous BAT-AELs ^a	Current BAT-AELs ^b
HCl	mg/Nm ³	10	2
SO ₂	mg/Nm ³	50	5

^a emission levels provided by Directive 2010/75/EU for waste incineration plants

^b lower end of the BAT-AEL range indicated by 2019 BREF WI (European Commission et al., 2020)

The higher efficiency in the removal of acid pollutants from WtE flue gas can be achieved applying a variety of techniques listed in the BATs, including the dry injection of different types of solid sorbents, the wet scrubbing with neutralizing solutions and combinations of dry and wet techniques. However, all the available techniques involve the consumption of chemical reactants and the generation of process waste, in the form of solid residues or wastewater streams, that require further treatment or disposal. As a consequence, the acid gas removal stage is typically the main driver of operating costs in WtE flue gas treatment lines (Dal Pozzo et al., 2016; Quicker et al., 2014) and it is responsible of a relevant share of indirect environmental impacts (the so-called cross-media effects), associated with the production of the reactants and the disposal of process waste.

Cross-media environmental impacts generally refer to a situation in which a reduction in the emission of a pollutant to one environmental medium results in increased emissions of other pollutants to the same or other environmental media (Rubin and McMichael, 1978). In the view of integrated pollution prevention, the EU framework for BAT determination takes into account at least qualitatively the issue of trans-medial problem shifting, explicitly considering the relevance of cross-media impacts as a criterion in the identification of BATs. More structured approaches for the quantification of cross-media effects based on life cycle assessment (LCA) and multi-criteria decision analysis (MCDA) have also been proposed to support technical working groups in the determination of BATs (Geldermann et al., 1999; European Commission, 2006; Geldermann and Rentz, 2008; Cakir et al., 2020).

However, the assessment of cross-media effects should be applied not only to the selection of BATs, but also to the selection of the level of performance at which BATs should be operated (i.e., the definition of ELVs at local, regional and national level). Actually, in a holistic framework, it should be ascertained that the environmental benefit due to the reduction of emission standards of a single pollutant is not offset by the associated negative impact deriving from the increase of cross-media effects. The possibility of burden shifting, i.e., the increase of the environmental impact of other life cycle stages involved in the process operations required to meet the new emission limits (Biganzoli et al., 2015), thus needs to be quantitatively assessed.

To date, relatively few studies were dedicated to the assessment of cross-media impacts correlated to the implementation of specific ELVs addressing a single environmental problem or compartment. Dong et al. (2021) and Phungrassami and Usubharatana (2021) focused on the technological retrofits needed to achieve ultra-low emissions in coal-fired power plants, evidencing a significant trade-off between the reduction of NO_x, SO_x and dust emissions at stack and the increase of impacts related to energy penalty and consumption of adsorbents. With respect to WtE facilities, Van Caneghem et al. (2016) showed that, if the European ELV for NO_x emissions would be halved, plants would be forced to replace SNCR deNO_x systems with tail-end SCR units and the net overall environmental impact could increase, particularly in the impact category global warming. Dal Pozzo et al. (2017) focused on dry technologies for acid gas removal and evidenced how lowering ELVs could disproportionately affect the indirect environmental impacts of single over multi-stage technologies. Dong et al. (2020) proposed an extensive evaluation of the impacts of different combinations of flue gas treatment technologies for MSW incinerators on the 18 impact categories of the ReCiPe method, showing that in 14 of them flue gas treatment generates additional burdens compared to a plant with untreated flue gas.

The present study addresses the quantification of cross-media effects considering the specific problem concerning the implementation of lower ELVs for acid gas emissions by WtE plants in the EU. A methodology is proposed for the quantitative assessment of cross-media impacts related to acid gas treatment, based on the combination of a simplified modelling of acid gas removal

processes with life cycle assessment (LCA). Starting from the increase of acid gas removal efficiency required by the introduction of lower emission standards, the additional reactant consumption and process waste generation are assessed using a portfolio of flue gas treatment process models allowing the performance simulation of the different acid gas removal technologies. The additional inputs and outputs of the acid gas removal process are then linked to a life cycle model that estimates the additional environmental impacts arising along the supply and disposal chain of reactants and waste streams. The quantitative assessment of the variation in the overall environmental impact is then displayed using a specific set of life cycle impact indicators. The additional overall flue gas treatment costs are also assessed, allowing a comparison of the environmental and economic dimensions. Italy was used as a case study to demonstrate the application of the methodology. The overall environmental and economic consequences of implementing lower acid gas emission limits and the extent of the environmental trade-offs and cross-media effects were assessed by the developed methodology.

2. Methodology

2.1 Overview

Assessing the cross-media effects caused by the introduction of lower emission standards for acid gas removal requires the quantification of the additional environmental impacts occurring in all the upstream and downstream activities involved in the operation of the specific WtE acid gas removal processes required to meet ELVs. Such impacts mostly derive from the increased consumption of reactants and from the increased generation of process residues, which in turn results in higher impacts related to their transportation and disposal.

Figure 1a reports the flow chart of the methodology developed to assess the relevant cross-media effects. In the following, the activities required to carry out the single steps of the methodology are discussed.

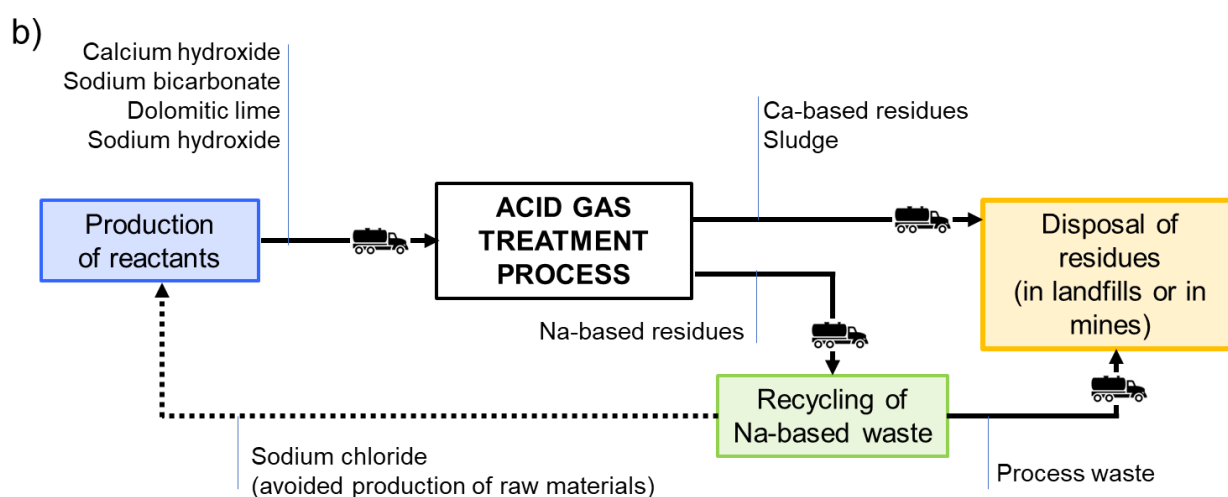
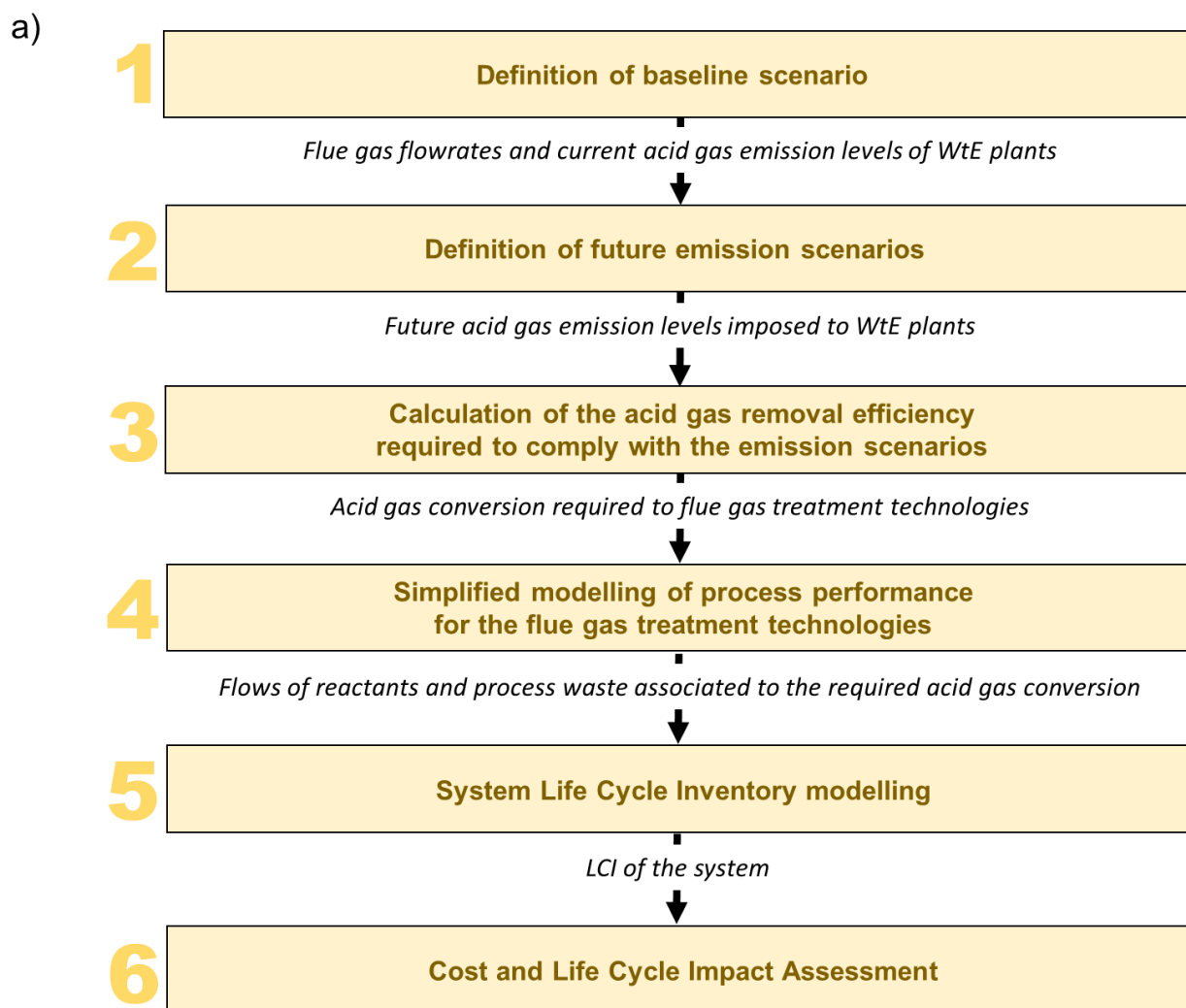


Figure 1. a) Flow chart of the methodology developed to assess cross-media effects deriving from the introduction of lower ELVs for acid gases in WtE facilities, b) generic LCA system boundary considered in the study. Technology-specific system boundaries are provided in section S2 of the SI.

2.2 Definition of baseline scenario

The first step in the procedure is the definition of the baseline scenario, i.e., the current emission levels by WtE plants (step 1 in **Figure 1a**). A reference area needs to be defined for the study (a region, a country, or several countries) where the same regulations apply. All WtE plants present in the area need to be identified and classified according to the acid gas treatment technology applied. For each plant, the current acid gas emissions need to be determined, typically by collecting the yearly averages of the concentrations of the acid pollutants (e.g., HCl and SO₂) in the flue gas at stack, $C_{i,out,t}$, and of the flue gas flowrate \dot{V}_{out} . These data can be retrieved, e.g., from the European Pollutant Release and Transfer Register (E-PRTR, 2021) for plants exceeding the reporting thresholds, from bulletins of the local environmental agencies, or from the environmental statements of plants operated by companies adopting the voluntary EMAS scheme.

2.3 Definition of future emission scenarios

In the second step of the procedure (see **Figure 1a**), the future emission scenarios are defined. Each future scenario is represented by a set of new ELVs for each acid pollutant considered in the study. The emissions of the WtE facilities considered need to be modified to comply with the changes assumed for the emission standards. More specifically, the definition of the setpoint of acid gases concentrations at stack that the plants will adopt has to consider a comfortable safety margin below the actual ELVs, as the plant operators will need to avoid that any deviation in process conditions (e.g., change in inlet acid gas load, operating temperature, reactant purity) will immediately cause exceeding the ELVs. In typical WtE flue gas treatment operation, setpoint concentrations at stack are usually set at 1/4 or 1/5 of the ELV (Dal Pozzo et al., 2021).

2.4 Calculation of the acid gas removal efficiency required to comply with the emission scenarios

In order to assess cross media effects, the conversion of acid gas during flue gas treatment needs to be assessed. In step 3 of the methodology (see **Figure 1a**), the acid gas removal efficiency

demanded to the flue gas treatment system in each emission scenario for the i -th acid pollutant considered, X_i , is calculated:

$$X_i = 1 - \frac{\dot{V}_{out} \cdot C_{i,out}}{\dot{V}_{in} \cdot C_{i,in}} \quad (1)$$

where \dot{V}_{in} and $C_{i,in}$ are the flue gas flowrate and the concentration of the acid pollutant i entering the flue gas treatment system. While it can be roughly assumed that $\dot{V}_{in} = \dot{V}_{out}$, neglecting the air infiltrations occurring along the flue gas treatment line, the concentration of acid pollutants (typically, HCl and SO₂) in the untreated flue gas is a critical parameter that needs to be assessed in order to quantify the required acid gas conversion. While data concerning the emission of pollutant at stack may be derived from relevant public documents by companies or regulatory agencies, as discussed in step 1, such documents seldom report the conversion of acid gas, the amount of acid gases removed in flue gas treatment and/or the amount of pollutants present in the untreated flue gas. Therefore, the typical flue gas composition upstream of the flue gas treatment system needs to be estimated. Considering that the release of pollutants such as HCl and SO₂ stems directly from Cl and S content in the waste, their concentration in untreated flue gas can be derived from a mass balance on the combustion process (e.g., see Dal Pozzo et al., 2016), given a reference waste composition for the type of waste feed (industrial or urban) or the location served by the WtE plant (Bisinella et al., 2022). Several plants measure the HCl and SO₂ concentration in the raw flue gas upstream of the flue gas cleaning line, e.g., at the boiler outlet, for process control purposes (Bacci di Capaci et al., 2022): if these data are made available, it is possible to directly derive average acid gas loads over a year. Alternatively, literature sources or technical data (European Commission et al., 2020) may provide relevant estimates. An example of the calculation of the conversion is provided in the discussion of the case study.

2.5 Simplified modelling of process performance for the flue gas treatment technologies

The following step (step 4 in **Figure 1a**) requires quantifying the consumption of reactants and the production of process waste related to flue gas treatment as a function of the required acid gas

conversion X_i , given by the amount of pollutants in the untreated gas (obtained in step 3) and the emission assumed at stack for each scenario (obtained in step 2):

$$[\dot{m}_{reactants}, \dot{m}_{process\ waste}] = f(X_i) \quad (2)$$

Clearly enough, the relationships summarized by Eq. (2) are specific for each process technology applied to acid gas removal. Thus, a simplified model needs to be selected or developed for each technology, allowing the assessment of technology-specific reactant consumption and waste production.

Currently, three main categories of process technologies are applied to acid gas removal: wet, semidry, or dry (Vehlow, 2015).

Wet acid gas treatment typically consists in a sequence of two scrubbers (Poggio and Grieco, 2010; Dal Pozzo and Cozzani, 2021): in the first column HCl is removed using water, while in the second column SO₂ and residual HCl are chemically absorbed using a water solution of sodium hydroxide (NaOH, 30% in weight). Wastewater produced by the scrubbers is treated adding lime for pH neutralization and, after sedimentation and thickening, sludges are sent to disposal.

In dry acid gas treatment technologies, powdered sorbents are injected in the flue gas stream to trigger gas-solid neutralization reactions with the acid gases and the solid reaction products are then removed using fabric filters. Sodium bicarbonate (NaHCO₃) and hydrated lime (Ca(OH)₂) are the most common reactants (Beylot et al., 2018; Liu et al., 2022). In recent years, a dry pre-treatment process based on the direct furnace injection of dolomitic lime (Ca(OH)₂·MgO, from the calcination and hydration of dolomite rock, see Biganzoli et al., 2015) has also found application in several WtE plants.

In semi-dry technologies, hydrated lime is mixed with water typically producing a 20-25% slurry by weight, which is then sprayed as atomized droplets to the flue gas in a specific scrubber (Ting et al., 2008; Yuan et al., 2022). Similarly to dry techniques, the semi-dry method generates solid process residues.

Several multi-stage configurations are applied in current industrial practice. In particular, two-stage dry treatment systems based on the combination of a 1st stage $\text{Ca}(\text{OH})_2$ injection and filtration and a 2nd stage NaHCO_3 injection and filtration have a widespread application (Dal Pozzo et al., 2018; De Greef et al., 2013). As mentioned above, the furnace injection of dolomitic lime can be considered as a pre-treatment stage and it is usually adopted in combination with downstream NaHCO_3 -based dry acid gas removal.

The features of multi-stage treatments allow the separate modelling of each stage of the treatment, provided that a repartition of acid gas removal between the different stages is defined. For a multi-stage treatment system with N stages, the overall conversion of the pollutant i , i.e., the removal efficiency required by Eq. (1), is given by:

$$X_{i,TOT} = \sum_{j=1}^N X_{i,j} - \prod_{j=1}^N X_{i,j} \quad (3)$$

where $X_{i,j}$ is the conversion of the pollutant i in the stage j .

Typically, the repartition of acid gas abatement between the different stages of the removal process is driven by economic considerations. Recent studies, focused on the economic optimization of multi-stage acid gas treatment systems (e.g., see Dal Pozzo et al. (2016) for hydrated lime + sodium bicarbonate systems and Dal Pozzo et al. (2020) for dolomitic lime + sodium bicarbonate systems), may be used to define reasonable values of the conversion in each stage, $X_{i,j}$, needed to set up the modelling of multi-stage treatment systems.

In the present study, simplified models as those proposed by Antonioni et al. (2014) were applied to describe the non-linear relationship between reactant feed rate and acid gas conversion in treatment systems. Section S1 of the Supporting Information (SI) provides details on the models selected and on their application to the case study.

Clearly enough, the proposed approach, summarized in **Figure 1a**, is not model-dependent and has a general validity. Thus, alternative models and/or modelling approaches, if available, may be applied as well to assess the performance of the acid gas removal technologies, provided that they

have sufficiently general features to allow their application to the different WtE facilities considered in the analysis.

2.6 System Life Cycle Inventory modelling

In step 5 of the methodology (see **Figure 1a**), the reference flows of reactants and process waste identified by the flue gas treatment process models are used to quantify the inputs (energy and material requirements) and the outputs (emissions) related to the supply and disposal chain, i.e., the life cycle inventory linked to the acid gas removal operation.

As outlined in **Figure 1b**, the system boundaries include the following life cycle stages:

- production of reactants and process water required by the acid gas removal system;
- operation of the acid gas removal system, according to the process models in step 4, including residual acid gas emissions at stack;
- operation of the wastewater treatment section, for wet systems;
- disposal of process residues as backfilling material for depleted salt mines or in hazardous landfill sites after stabilization;
- recycling of process residues, for bicarbonate-based residues;
- all the transportation phases among the aforementioned processes.

The system boundaries and the life cycle modelling associated with the operation of each acid gas treatment technology are detailed in section S2 of the SI. In this study, the nodes of the Life Cycle Data Network (European Commission, 2014) were used as main source of data, complemented with literature sources and appropriate assumptions where needed.

For the production of reactants, the entire supply chain is taken into account, from the extraction and production of raw materials and base reactants to the transformation processes that deliver the final reactants, with the associated consumption of electricity, heat, water, and fuels. Hydrated and dolomitic lime are mining products, deriving from the quarrying of limestone and dolomite rock, respectively, and their subsequent grinding, calcination and slaking in water. Sodium bicarbonate is a chemical product, obtained from the hydration and carbonation of sodium carbonate (Pacher et

al., 2009), which in turn is produced from sodium chloride, ammonia, and calcium carbonate through the Solvay process (Bonfim-Rocha et al., 2020). Sodium hydroxide is produced by electrolysis of sodium chloride solutions in the chlor-alkali process (Garcia-Herrero et al., 2017).

The operation of the acid gas treatment technologies is modelled according to the models introduced in step 4 of the methodology. The direct impacts of this phase include the residual acid gas emissions at stack. In addition, environmental consequences from the decomposition of reactants or their interaction with other pollutants should be taken into account. In particular, once injected in the flue gas, hydrated lime can absorb carbon dioxide through the reaction (Chin et al., 2005):



In the context of acid gas removal, this is an undesired reaction, as it consumes part of the reactant feed rate injected for acid gas neutralization. However, the modest positive side effect of CO₂ capture should be accounted. A quantification of the mass of absorbed CO₂ per mass of injected reactant can be derived from process residues composition data (see, e.g., Dal Pozzo et al., 2018b), as detailed in section S2 of the SI. In the present study, it was assumed that on average 1 kg of Ca-based residues contains 0.13 kg of CO₂. Conversely, at the typical temperatures of the flue gas treatment system (150-200 °C, Dal Pozzo et al., 2019), sodium bicarbonate decomposes entirely to sodium carbonate releasing CO₂ and water:



Thus, 0.52 kg of CO₂ are emitted for each kg of NaHCO₃ injected in the flue gas.

The process residues formed in the acid gas removal operation, either sludges from the wastewater treatment of the effluents of wet scrubbers or solid residues from dry and semi-dry techniques, are typically sent to disposal. These residues streams are frequently mixed with fly ash, activated charcoal, trace metals and other absorbed micro-pollutants (Bogush et al., 2015; Margallo et al., 2015), hence labelled as hazardous waste with very limited commercial valorization routes available to date (Nedkvitne et al., 2021; Quina et al., 2020). Two disposal strategies are thus available: i) disposal in hazardous waste landfills; or ii) disposal as backfilling material in exhausted salt mines.

In the first case, an inertization pre-treatment (mixing with cement) is also considered to reduce the leaching potential of the disposed residues. An alternative valorization route is available only for the Na-based process residues stemming from sodium bicarbonate dry injection. If these residues are collected separately from fly ash (e.g., in two-stage dry systems or in single stage systems with upstream fly ash removal), they can be sent to dedicated chemical recycling plants that recover a sodium chloride brine suitable as raw material in the production of new sodium bicarbonate, minimizing the residual fraction destined to disposal (Brivio et al., 2018). Here, the three alternatives for the management of process residues were modelled focusing on the material and energy demand of the disposal or valorization routes.

Another potentially relevant environmental impact is directly related to the long-term leaching of metals and organic contaminants from the residues after disposal (Luo et al., 2019). However, this impact is mainly associated to the fly ash and activated charcoal streams that are collected together with the process residues of acid gas removal: as such, it is present both in the current and in the future scenarios, and it is largely independent of the amount of acid gas removal reactants fed to the system. Therefore, for the sake of simplicity, in the framework of a differential analysis between acid gas removal scenarios, the potential impacts related to landfill leaching were excluded from the analysis, as they were supposed not to be influenced by changes in operating conditions across scenarios.

Construction and installation of the acid gas treatment technologies were excluded from the boundaries of the case study introduced in section 3, since it can be reasonably assumed that currently installed technologies have the potential to meet the future emission standards by a modification of the operating conditions, with no need to modify the process equipment, provided that the totality of the WtE plants considered already apply BATs for acid gas removal (see, e.g., Ardolino et al., 2020).

2.7 Cost and Life Cycle Impact Assessment

Once the full inventory of the system has been quantified under the baseline and the future scenarios, it is possible to proceed with the life cycle impact assessment (LCIA). Four midpoint

impact categories were taken into account: resource depletion with reference to fossil fuels (RD, expressed in MJ), global warming (GW, expressed in $\text{kg}_{\text{CO}_2, \text{eq.}}$), acidification (AC, expressed in $\text{kg}_{\text{SO}_2, \text{eq.}}$), and smog formation (SF, expressed in $\text{kg}_{\text{ethylene, eq.}}$). The characterization factors provided by the CML-IA database were adopted (Guinée et al., 2002). The methodology may in principle support different LCIA approaches and more ample sets of environmental indicators. However, in order to limit the complexity of the results, the analysis only considered the four environmental categories that previous studies on the life cycle impacts of acid gas treatment technologies identified as the most critically affected (Ardolino et al., 2020; Dal Pozzo et al., 2017; Dong et al., 2020; Scipioni et al., 2009).

In order to provide a direct comparison across impact categories and to allow for the aggregation of the results, normalization can also be applied. External normalization was adopted in the case study, dividing the scores of the impact categories by normalization factors derived from the work by Crenna et al. (2019) on global emissions and resource uses, as detailed in section S3 of the SI. As discussed in section 4, normalization can help in the quantitative assessment of trade-offs between impact categories.

Eventually, in order to contemplate also the trade-off between the environmental and the economic dimensions (Chen et al., 2022), step 6 of the methodology addresses also the quantification of the additional costs required to meet the future emission scenarios defined in step 2 (see **Figure 1a**). The operating costs of the acid gas treatment process are estimated on the basis of the unit costs for the purchase of reactants and the management of process residues. As for capital costs, in general, additional costs due to the introduction of new or improved flue gas treatment processes required to meet the new emission limits should be considered, but – as illustrated in section 2.6 – it can be assumed that no modifications in process layout are needed to comply with the future emission scenarios considered in the case study.

3. Case study

3.1 The Italian municipal solid waste incineration sector

The municipal solid waste incineration (MSWI) sector in Italy was selected as a case study for the application of the methodology presented above. Italy is a member state of the European Union, thus applying the IED and BREF-based environmental permitting.

Data on the MSWI facilities operating in Italy, on their acid gas treatment technologies and on their current emission levels were obtained from reports of the regional environmental agencies, from environmental statements of the WtE companies and from direct data exchange with plant managers. Based on the documentation retrieved, 36 MSWI WtE plants resulted currently operating in Italy and were included in the analysis. **Figure 2** shows the location of the WtE plants, specifying the acid gas treatment technology applied and the size in terms of waste treatment capacity. A summary of the detailed data collected for each WtE facility, concerning the amount and type of waste streams incinerated per year, the acid gas treatment technology and the current emission levels of HCl and SO₂, is provided in section S4 of the SI.

As shown in **Fig. 2**, most of the Italian WtE plants are limited in size. However, in the reference year considered, 54% of the total incinerated wastes are treated in the 7 large-scale WtE facilities. The technologies for acid gas treatment adopted in Italian WtE plants can be classified in 8 categories, as summarized in **Table 2**. A total of 15 WtE plants apply single-stage removal techniques, based on dry or semi-dry injection of hydrated lime (technologies T1 and T2) or of sodium bicarbonate (T3). Other 15 WtE facilities adopt two-stage dry techniques, based on the combination of hydrated lime and sodium bicarbonate (T4), or dolomitic lime and sodium bicarbonate (T5). The remaining plants apply wet treatment systems: 3 facilities rely exclusively on wet scrubbers (T6), while the others use wet scrubbing as the final stage, after a dry treatment with lime (T7) or furnace injection of dolomitic lime and dry treatment with sodium bicarbonate (T8). The simplified process schemes of the technologies described in **Table 2** are provided in section S2 of the SI.

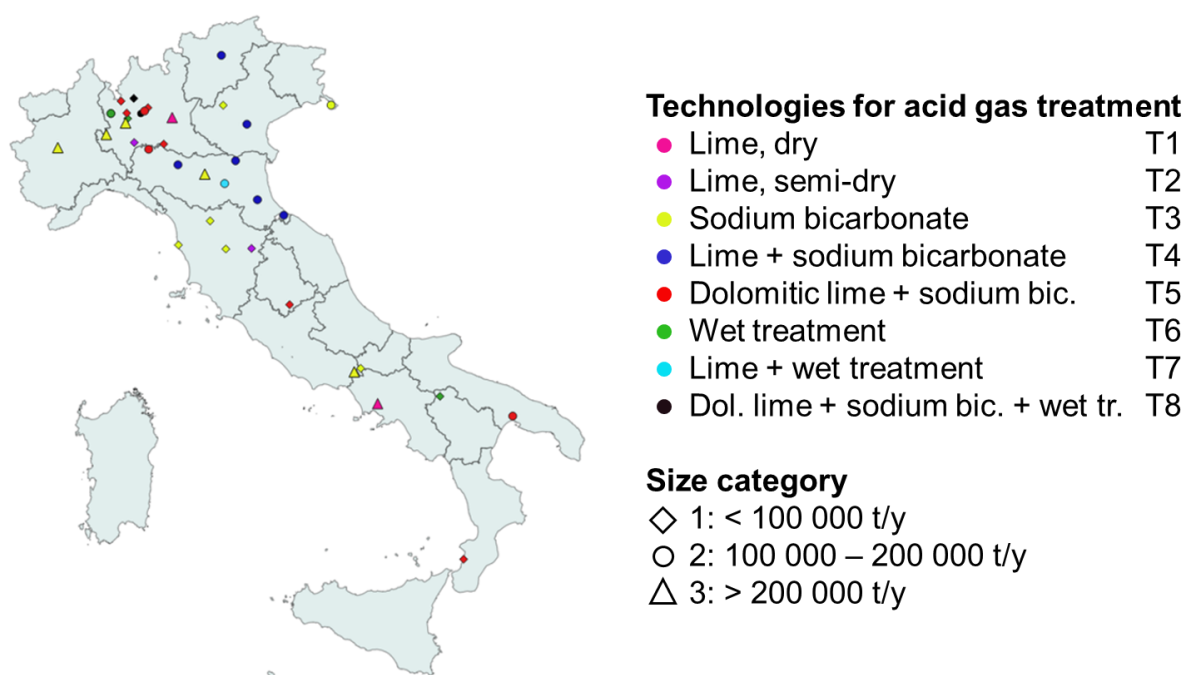


Figure 2. Map of Italian WtE plants classified according to size and acid gas treatment technology. See Table 2 for the description of technologies for acid gas treatment (T1 – T8).

Table 2. Types of acid gas treatment technologies adopted in the Italian WtE plants. Reference schemes are provided in section S2 of the SI.

Tech. ID n°	Acid gas treatment technology	Description
T1	Lime, dry	Injection of dry powdered $\text{Ca}(\text{OH})_2$ in the flue gas duct, followed by separation of reaction products via a fabric filter. High surface area (HSA) lime is typically used in this technique.
T2	Lime, semi-dry	Injection of an atomized slurry of water and $\text{Ca}(\text{OH})_2$ 20-25% in a scrubber, followed by separation of reaction products via a fabric filter.
T3	Sodium bicarbonate	Injection of dry powdered NaHCO_3 in the flue gas duct, followed by separation of reaction products via a fabric filter.
T4	Lime + sodium bicarbonate	Injection of dry powdered $\text{Ca}(\text{OH})_2$ followed by the injection of dry powdered NaHCO_3 . After each step, a fabric filter collects the respective reaction products.
T5	Dolomitic lime + sodium bicarbonate	Injection of dolomitic lime directly in furnace, followed by injection of dry powdered NaHCO_3 in the flue gas duct. Dolomitic residues are collected at the bottom of the boiler, while Na-based residues are separated via a fabric filter.
T6	Wet treatment	Sequence of two scrubbers: in the first column, water is provided, while in the second column an aqueous solution of NaOH 30% in weight is used. The effluents are sent to a dedicated wastewater treatment.
T7	Lime + wet treatment	Injection of dry powdered $\text{Ca}(\text{OH})_2$ in the flue gas duct, followed by separation of reaction products via a fabric filter and by a two-scrubber system (water + NaOH solution) with the related wastewater treatment.
T8	Dolomitic lime + sodium bicarbonate + wet treatment	Injection of dolomitic lime directly in furnace, followed by injection of dry powdered NaHCO_3 in the flue gas duct. Dolomitic residues are collected at the bottom of the boiler, while Na-based residues are separated via a fabric filter. The last step is a two-scrubber system (water + NaOH solution) with the related wastewater treatment.

3.2 Application of the methodology to the case study

In this section, the application of the methodology described in section 2 to the case study is illustrated, and the assumptions introduced are discussed. The analysis is performed considering a year of operation of the WtE plants. The baseline scenario is represented by the current HCl and SO₂ emissions in the 36 WtE plants. In order to define the scenario for the case study, the average HCl and SO₂ concentrations at stack in the last year of available data were taken for each plant considered as representative of the current emission level of the facility. Where not available, the total flue gas flowrate of the plant was estimated from the amount of waste incinerated considering a wet flue gas to waste ratio of 6 Nm³/kg waste (IPCC, 2001).

With respect to the definition of the future scenarios, it is assumed that the local authorities will set future ELVs based on the lower end of the BAT-AEL range reported in **Table 1**, i.e., 2 mg/Nm³ for HCl. Considering that plant operation should ensure a safety margin from the ELVs (as discussed in section 2.3), a set of 10 future emission scenarios was defined in the study, assuming that all the WtE plants need to maintain a fixed setpoint for HCl concentration at stack ranging from 2 to 0.2 mg/Nm³ according to the scenario (see **Table 3**). The range of emission levels explored by the scenarios in **Table 3** is a reasonable assumption. The current yearly HCl concentration at stack has an average of 2.00 mg/Nm³ for the 36 MSWI facilities in Italy and a standard deviation across plants of 1.45 mg/Nm³. A renewal of the environmental permit in the view of the new BREF WI would likely issue lower ELVs. Notably, a few plants located in critical areas (heavily industrialized districts in territories with unfavourable conditions for atmospheric dispersion) already operate with emission setpoints near the lowest levels defined in **Table 3**. Thus, for each scenario in **Table 3**, if the current emission level is already lower than the new setpoint of concentration at stack, the current lower emission level is maintained also in the new scenario. If the current average value of HCl concentration at stack exceeds the setpoint of concentration of the new scenario, compliance with the new value is considered.

It is worth noting that **Table 3** only reports reference values for HCl emissions. Considering that HCl is typically present in untreated flue gas from MSWI in concentrations at least 5-10 times higher than

SO₂, HCl is the critical pollutant in terms of emission compliance. If the plant is able to comply with the HCl setpoints in **Table 3**, the SO₂ concentration at stack calculated by the process models for the different acid gas removal technologies in **Table 2** results always lower than the lower end of the BAT-AEL range in **Table 1** (5 mg/Nm³).

Table 3. HCl emission setpoints considered for the baseline and future scenarios.

Scenario	Emission setpoint for HCl (mg/Nm ³)
S0	Baseline scenario (current average HCl concentration at stack in the last year of data available)
S1	2
S2	1.8
S3	1.6
S4	1.4
S5	1.2
S6	1
S7	0.8
S8	0.6
S9	0.4
S10	0.2

As discussed in section 2.4, in order to quantify the acid gas conversion of the acid gas removal system and to calculate the related flows of reactants and process waste, the composition of the untreated flue gas is required. Ideally, the average concentration of HCl and SO₂ in the untreated flue gas upstream of the flue gas cleaning line should be known for each WtE plant.

Actually, as mentioned in section 2.4, this information is seldom reported in public data. Here, reference values for HCl and SO₂ concentration were adopted to represent the average composition of untreated flue gas in all the WtE plants considered in the case study, depending on the type of waste feed: for plants treating municipal solid waste, HCl and SO₂ concentrations in the raw gas were assumed equal to 800 and 100 mg/Nm³, respectively, while, for plants treating mixed urban

and industrial waste, they were assumed equal to 1000 and 200 mg/Nm³, respectively. These values were derived from literature sources on typical MSWI flue gas compositions and from operational data of a few Italian plants among those included in **Figure 2**. Section S5 of the SI provides an example of the time series analysed to obtain the reference data reported.

In order to check that this simplifying assumption does not introduce unacceptable errors in the estimate of the mass flows of reactants and process residues associated with the acid gas removal operation, a validation was performed for a few plants for which data of current reactant consumption were available from their EMAS statements. For each plant, the annual mass of reactants reported in the EMAS document was compared with the mass calculated from the process model considering the baseline scenario and the aforementioned values of HCl and SO₂ as the input flue gas composition. The maximum deviation among the actual and the calculated amounts of reactants was always lower than 11%. The validation procedure and results are discussed in section S5 of the SI.

The models introduced in section 2.5 and detailed in section S1 of the SI were used to describe the acid gas removal process for each technology or technology combination.

In the case of multi-stage systems, the following assumptions on the repartition of abatement between stages were adopted. For two-stage lime + bicarbonate systems (T4), a HCl conversion equal to 70% in the 1st hydrated lime-fed treatment stage was assumed, as this is a typical value that minimizes the overall operating cost of the two-stage system (Dal Pozzo et al., 2016). Analogously, for two-stage dolomitic lime + bicarbonate (T5), the furnace injection of dolomitic lime was assumed to achieve a 10% HCl conversion (Biganzoli et al., 2015; Dal Pozzo et al., 2020).

The life cycle inventories for each WtE plant were modelled as discussed in section 2.6 and in section S2 in the SI. A distance of 300 km was considered as a realistic estimate for the transportation of reactants. With respect to the fate of process wastes, considering that the disposal of process residues as backfilling material in depleted underground salt mines is usually performed in Germany (Frøer et al., 2010), it was assumed that only plants located in the North of Italy adopt this disposal route for their residues, while plants in the Center and in the South of Italy resort to stabilization and disposal in hazardous waste landfills. As a consequence, transportation distances

were set at 800 km for salt mine disposal and 300 km for landfill disposal. A distance of 300 km was considered also for the recycling plants dedicated to Na-based residues.

The unit costs reported in section S6 of the SI, obtained considering data provided by several process managers of Italian WtE facilities and relevant technical literature, were considered for the cost entries included in the assessment.

4. Results and discussion

4.1 Global environmental impacts for the Italian MSWI sector

The effects of the transition from the current baseline scenario S0 to the future emission scenarios (S1-S10 in **Table 3**) on the global mass balance of acid gas removal in Italian MSWI WtE plants are shown in **Figure 3**, that reports the % variation in the mass flows of acid gas emissions, reactant consumption and process waste generation. Absolute figures for each scenario are reported in section S7 of the SI.

On the one hand, the application of the stricter future emission scenario (S10) results respectively in a 91% and a 70% reduction of the total mass flows of HCl and SO₂ emitted by Italian WtE plants with respect to the current (baseline) scenario. The difference among HCl and SO₂ is due to the lower reactivity of the main reactants adopted in acid gas removal technologies towards SO₂.

On the other hand, the consumption of chemicals in the acid gas removal section of WtE facilities is expected to increase: in scenario S10, the demand of Ca(OH)₂ and NaHCO₃, the main reactants adopted in dry and semi-dry technologies, is respectively 18% and 22% higher than in the baseline scenario. The mass flows related to wet acid gas abatement, i.e., the make-up of process water and the consumption of NaOH, exhibit significantly lower rises (0.04% and 1.17%, respectively). The total mass flow of process waste from dry, semi-dry and wet treatment systems increases by 15% when the emission scenario changes from S0 to S10.

The reference material flows analysed in **Figure 3** are the basis for the quantification of the life cycle impacts related to acid gas removal. On the basis of the life cycle inventory modelling outlined in section 2.6 and the impact assessment approach discussed in section 2.7, the life cycle impacts for the sum of the Italian WtE plants on the four selected impact indicators are reported in **Figure 4**. The contribution of the life cycle stages to the overall impacts is also highlighted in the figure.

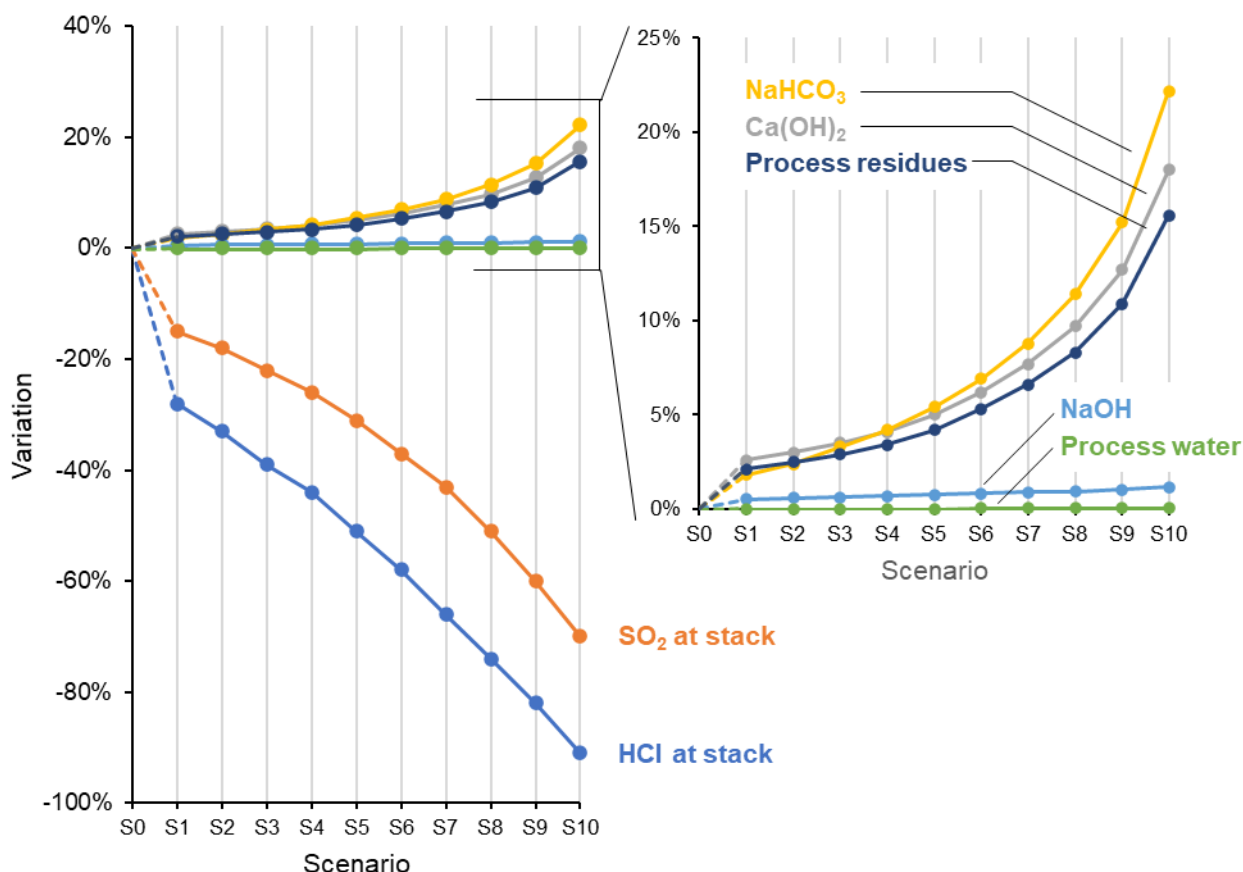


Figure 3. Variation in the mass flows of acid gas emissions, reactant consumption, and process waste generation as a function of the emission scenarios (S0 – S10) introduced in Table 3, for all the WtE facilities considered in the case study. Emission at stack = after treatment.

Figure 4 shows that moving from the baseline scenario, S0, to the stricter future emission scenario, S10, impacts in the acidification category decrease (-22% from S0 to S10) as expected, due to the reduction of total burdens achieved by lower emission standards comes as a balance between lower direct impacts and higher indirect impacts. However, impacts in the resource depletion and global warming categories increase (+20% from S0 to S10), and impacts in the smog formation category remain relatively unchanged (+1.2% from S0 to S10).

With respect to acidification (**Fig. 4a**), the total acidification in the baseline scenario is estimated at 3.48×10^5 kg_{SO₂,eq./y}. The reduction of HCl and SO₂ emissions at the stack of WtE plants corresponds to a reduction of up to 1.19×10^5 kg_{SO₂,eq./y} (scenario S10) in the acidification impact. However, the environmental benefit at the WtE plants is compensated by the increase of indirect impacts along

the supply and disposal chain, hence producing a net acidification reduction of $7.68 \times 10^4 \text{ kg}_{\text{SO}_2, \text{eq.}}/\text{y}$ for scenario S10.

The partial life cycle burden shifting across scenarios is evident: while the fraction of impacts associated with direct HCl and SO₂ emissions at the plants decreases from 42% in emission scenario S0 to 11% in scenario S10, the share related to reactant production, which is the main contributor to life cycle acidification impacts, increases from 52% to 81%. These burdens are at least partially compensated by the avoided emissions associated with the recycling of Na-based residues, which substitutes virgin raw materials for NaHCO₃ production, providing an associated benefit of $4.10 \times 10^4 \text{ kg}_{\text{SO}_2, \text{eq.}}/\text{y}$ in scenario S10. To provide a meaningful term of comparison for the acidification reduction granted by the strictest emission scenario S10, it is worth noting that the total Italian emission of SO₂ as reported by E-PRTR for the reference year 2019 is $1.05 \times 10^8 \text{ kg}$.

Lowering the HCl emissions, the resource depletion indicator (**Fig. 4b**) shows increasing values, as the intensification of the acid gas removal operation entails a higher consumption of energy and materials along the supply and disposal chain. A 20% increase in the score of the indicator is estimated when the emission scenario changes from S0 to S10. An even higher share of burdens than in the acidification category (86% for S10) is associated with the production of reactants, which is the most energy-intensive life cycle stage.

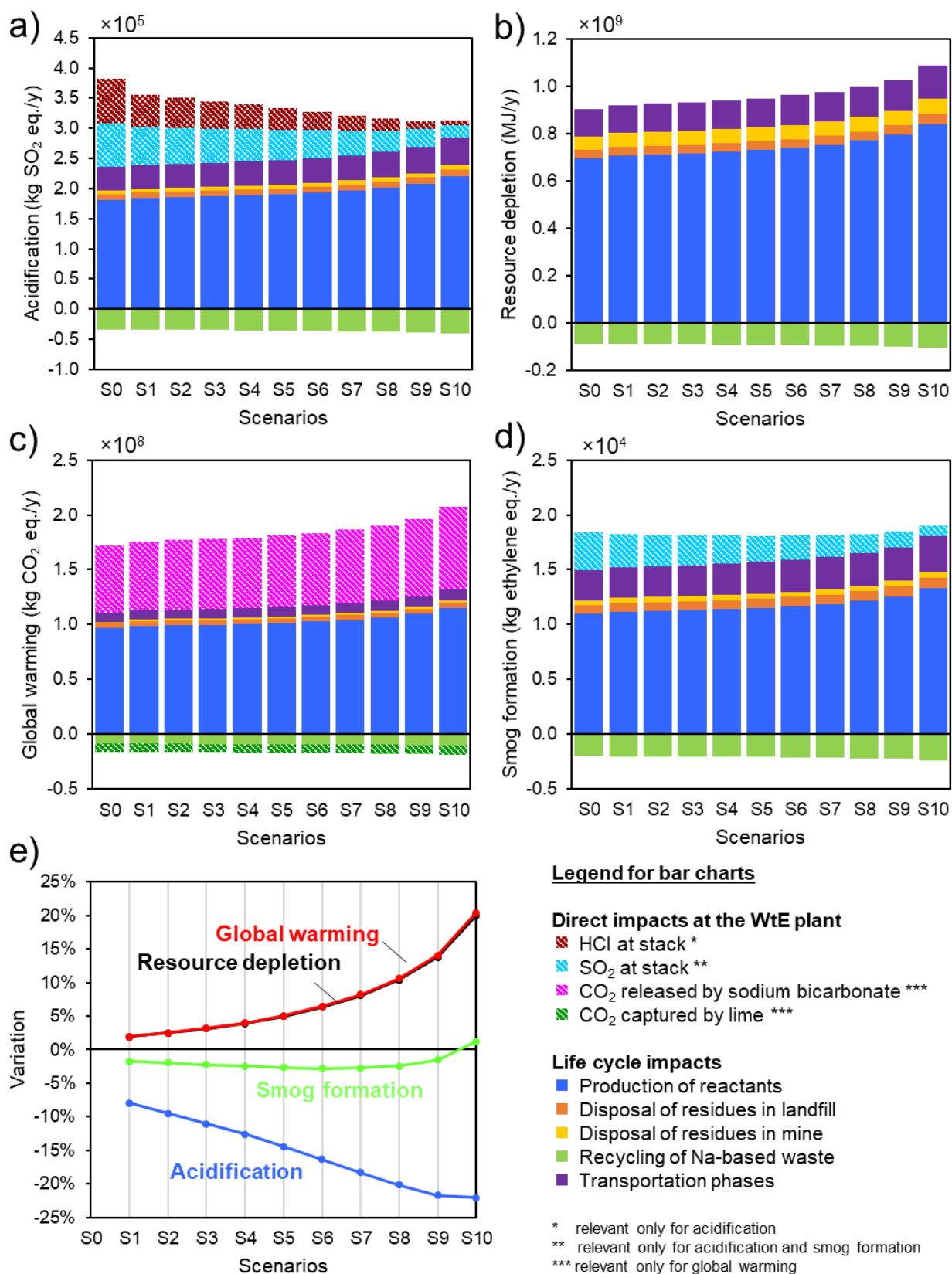


Figure 4. Values calculated for the selected environmental impact indicators in the emission scenarios considered: a) acidification, b) resource depletion, c) global warming, d) smog formation. e) % variation of the overall score of the four indicators across scenarios. Emission scenarios (S0 – S10) are described in **Table 3**.

The production of reactants is the main contributor also to the global warming (GW) impact category (**Fig. 4c**), although a relevant share of burdens stems directly from the emissions at the WtE plants. The release of CO₂ by NaHCO₃ through reaction (5) accounts for 40% of the total GW impacts (S10). while the CO₂ absorption by Ca(OH)₂ provides avoided GHG emissions equal to 5% of the S10 GW impacts. Altogether, the GW indicator increases by 20% when the emission scenario is changed between S0 and S10.

Lastly, the smog formation category (**Fig. 4d**) shows relatively stable impacts across the emission scenarios, with a difference of less than 1.2% between S0 and S10. The overall effect comes as a compensation between the reduction of direct emissions of SO₂ at the WtE stacks and the increase of life cycle burdens. Reactant production is confirmed as the main contributor to the latter, while the transportation phases add up to a relatively higher share of impacts than in the other categories (20% in S10), owing to the release of NO_x and VOCs by trucks.

Fig. 4e summarizes the global outcome of the new emission scenarios for the four environmental categories considered, reporting the % variation of the impact indicators with respect to the baseline scenario. The figure clearly evidences the trade-off between environmental impacts associated with lower acid gas ELVs at the WtE stacks.

Figure 5 provides an overall evaluation of the consequences of the new emission scenarios on the impact categories under study, obtained normalizing and piling the four indicators in a bar chart. The external normalization approach introduced in section 2.7 was applied. It should be remarked that, as already mentioned in section 2.7, the set of indicators adopted in this study does not aim to be exhaustive, since other impact categories are potentially affected by the system under study. Therefore, the results reported in **Figure 5** only provide a rough representation of the overall impact, useful in the framework of the discussion that follows, but are not intended to be used as a holistic environmental index. Moreover, no weighting between the indicators was performed, to avoid introducing a subjective element in the analysis. On the one hand, among the impact categories considered, most existing LCIA weighting schemes, often based on stakeholder surveys, assign greater relevance to global warming (Sala et al., 2018). On the other hand, local policies, together

with the new ELVs, may introduce priorities addressing, e.g., the reduction of impacts on a local or regional scale (acidification, smog formation) over impacts on a global scale (global warming, resource depletion).

Nonetheless, it is worth noting that the sum of the normalized indicators reported in **Figure 5** evidences that stricter limits in acid gas emission scenarios are not associated with a clear overall environmental benefit. Actually, the reduction in the acidification impact appears more than offset by the increased burdens in the global warming and resource depletion categories when moving from emission scenario S0 to S10.

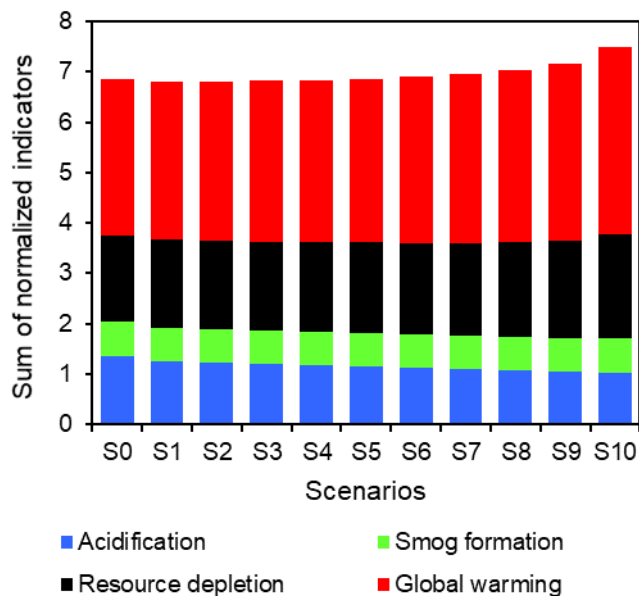


Figure 5. Values of the normalized indicators calculated for the case study. **Table 3** reports the description of the emission scenarios considered (S0 – S10).

The numerical results reported above are sensible to the assumptions introduced in the analysis. Two main sources of uncertainty are present: i) the modelling of flue gas treatment performance, ii) the modelling of the life cycle inventories. Both aspects were explored in a specific sensitivity analysis, reported in section S8 of the SI. The observed trends of the environmental indicators across scenarios are robust and were confirmed by the results of the sensitivity analysis.

4.2 Environmental impacts of the WtE facilities considered in the case study

The global results of section 4.1 can be broken down to assess the contributions of the individual WtE facilities considered in the case study. **Figure 6** shows the life cycle impacts for each plant in the acidification category in the baseline scenario, S0 (panel a), and in the stricter emission scenario, S10 (panel b).

It can be noticed that the direct impacts related to HCl and SO₂ releases at WtE stacks strongly decrease when the emission scenario is modified from S0 to S10, while the contributions related to life cycle emissions increase. Clearly enough, environmental burdens depend on the size of the plants and the 7 plants in the highest size category of Fig. 2 originate the 56% of the global impact. However, interesting technology-related patterns can be observed, also considering the results per unit mass of waste treated reported in section S7 of the SI. In particular, plants equipped with dry treatment systems exhibit the highest impacts and require the highest increase of life cycle impacts to comply with the reduced HCl and SO₂ emissions at stack in scenario S10. For bicarbonate-based single-stage systems (technology T3), the increase of impacts is particularly relevant for reactant production, as the NaHCO₃ supply chain involves energy-intensive processes that entail acid gas emissions for electricity and heat requirements.

Fig. 6c shows the variation of three indicators (acidification, smog formation, and global warming) in scenario S10 with respect to the baseline scenario S0 in each plant and further clarifies how trade-offs between environmental categories are technology-related. In particular, the figure shows that the increase in the global warming indicator associated with stricter emission scenarios (Fig. 5c) is mainly caused by lime- and bicarbonate-based single-stage systems (technologies T1, T2, and T3). Plants equipped with lime-based systems realize a 47% acidification reduction when moving to S0 to S10, but their global warming impact increases by 39%, mainly due to the CO₂ release associated with lime production (calcination of limestone) that is only minimally offset by the subsequent small CO₂ uptake by lime, once injected in the WtE flue gas. Plants equipped with bicarbonate-based systems achieve a 13% reduction in the acidification indicator when considering the S10 emission scenario with respect to the baseline S0 scenario, but their global warming impact increases by 24%,

mainly owing to CO₂ emissions in the bicarbonate production process and CO₂ release when the reactant is injected in the WtE hot flue gas.

Conversely, two-stage systems that couple a 1st lime-based stage with a 2nd bicarbonate-based stage (technology T4) optimize reactant consumption and are thus able to reduce the global warming cross-media effect: the 32% acidification reduction achieved when implementing the S10 emission scenario is obtained with only a 5% increase in global warming burdens. Wet treatment systems (T6) achieve a 29% reduction in the acidification indicator with a negligible increase in global warming burdens (+0.10%).

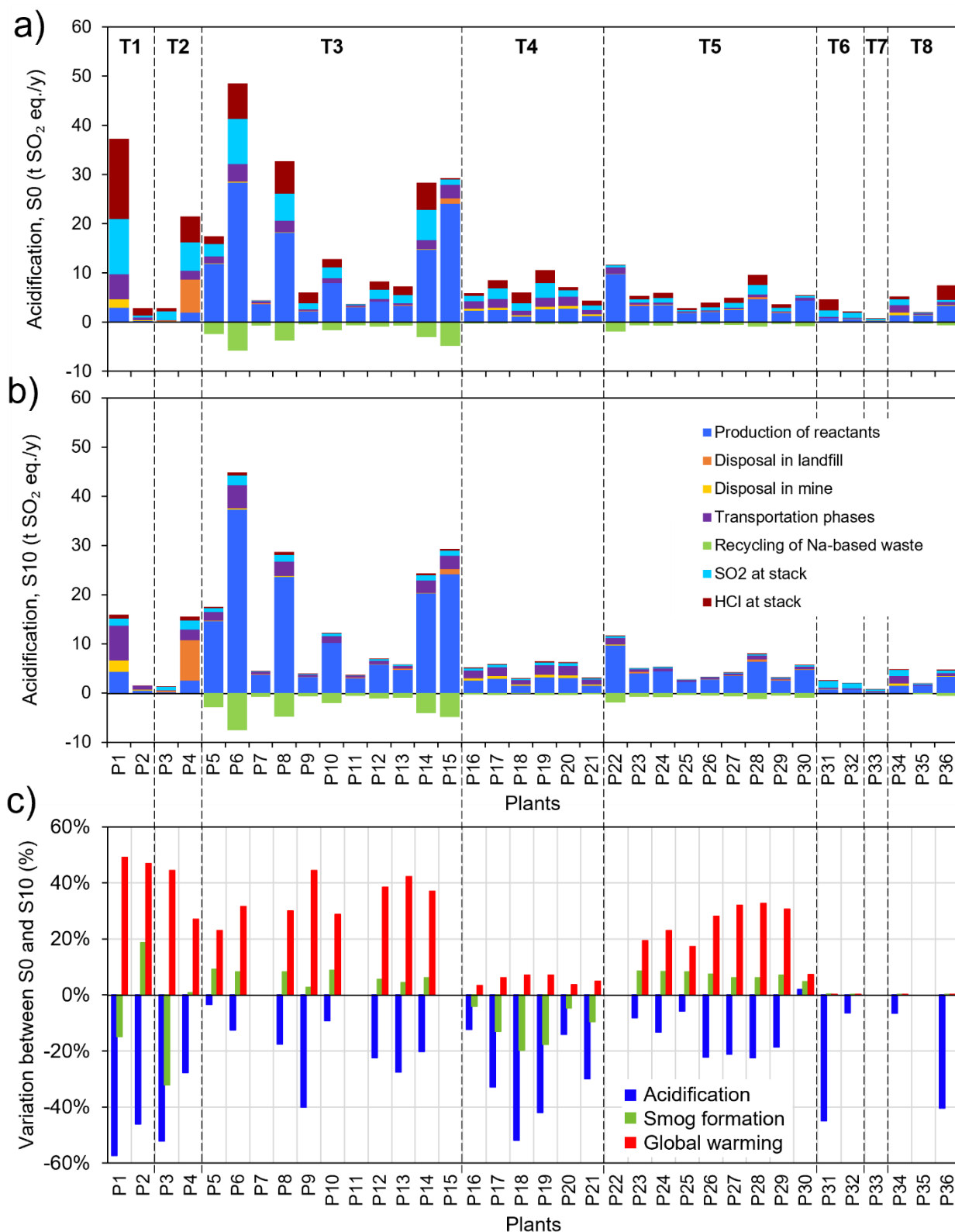


Figure 6. Acidification impacts for each plant in: a) baseline scenario S0, b) scenario S10. Plants are grouped considering the acid gas removal technology (T1 – T8: acid gas removal technology; **Table 2** reports the description of each technology. P1 – P36: plant identification tag; **Table S12** in the SI reports the more relevant data of each plant considered).

4.3 Economic and environmental trade-offs

Figure 7 shows the overall operating costs of acid gas abatement for the different emission scenarios considered in the case study, calculated on the basis of the unit costs reported in section S6 in the SI. As mentioned in section 2.7, only operating costs related to the procurement of reactants and the management of process waste are considered in the case study. **Figure 7a** highlights that costs increase from scenario S0 to S10, driven by the growing demand of reactants and the subsequent need to manage their process residues. In all the scenarios, 55% of the overall operating cost is due to the cost of reactants, 25% is due to the recycling of sodium-based waste and 20% to the disposal of residues. When implementing scenario S10, a 21% increase in the total operating costs with respect to baseline scenario is recorded.

As shown in **Figure 7b**, the increase in costs is not linear with the amount of acid gases removed in the plants. As expected, a higher unit operating cost per kg of acid gas removed is obtained when lower emission limits are implemented, since the use of more severe conditions and of higher excess of reactants are needed to remove low concentrations of pollutants in the flue gas.

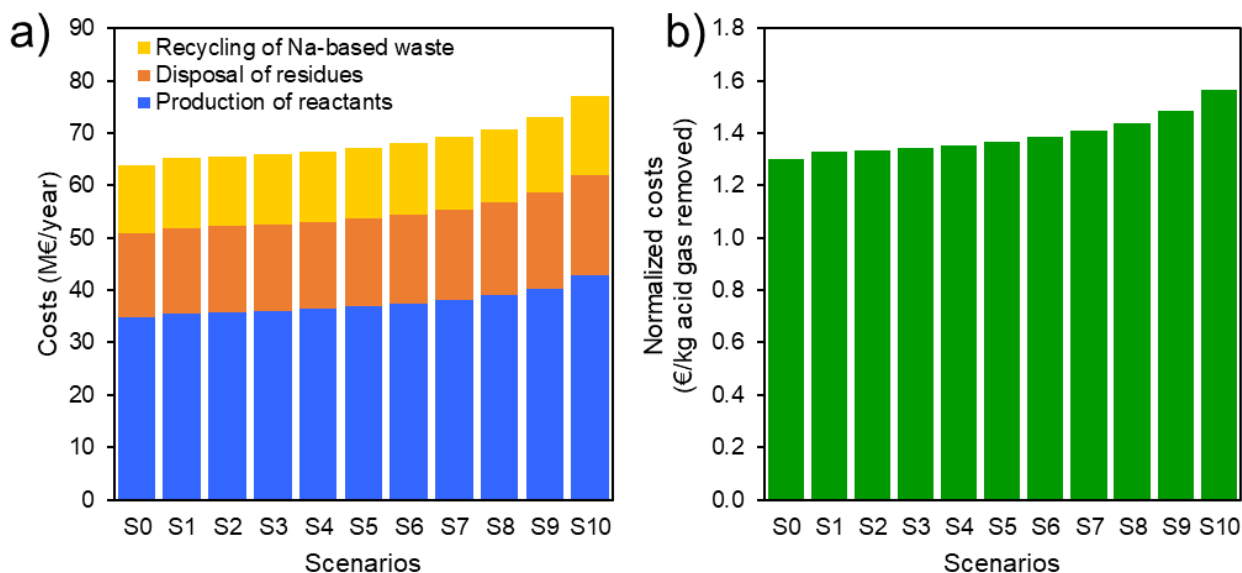


Figure 7. Cost assessment for the sum of Italian WtE plants as a function of emission scenario: a) total costs and cost contributions, and b) specific costs per unit of acid gas removed expressed as SO₂ equivalent. Emission scenarios considered (S0-S10) are described in **Table 3**.

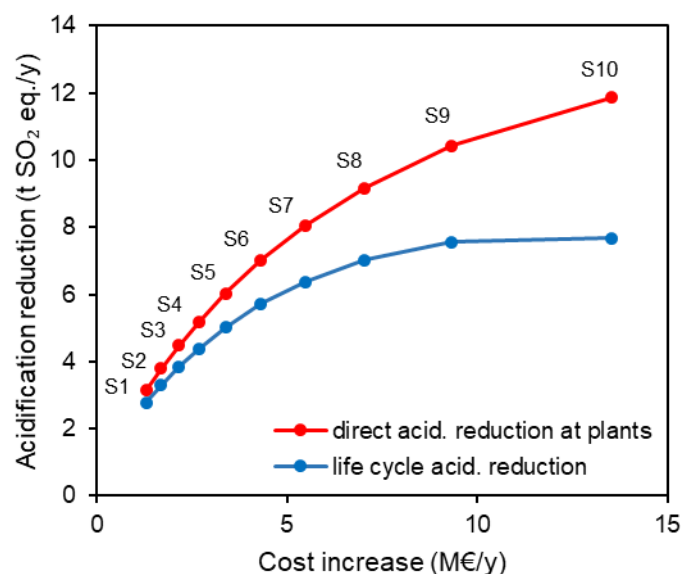


Figure 8. Direct acidification reduction at the WtE stacks and net acidification reduction considering the entire life cycle with respect to the increase in acid gas removal operating costs calculated for the emission scenarios considered in the case study. Emission scenarios considered (S0-S10) are described in **Table 3**.

The results shown in **Figure 7** point out that the imposition of lower ELVs implies a trade-off between the economic and the environmental dimensions. To better quantify the relationship between the reduction of HCl and SO₂ emissions at stack and the related costs, **Figure 8** reports the reduction of the acidification impact in each scenario with respect to the increase of abatement cost for the WtE acid gas removal systems compared to the baseline scenario. Both the reduction of acidification impact occurring at the stacks of the WtE plants (red curve, only considering direct emissions of HCl and SO₂) and the net reduction in the acidification indicator considering the whole life cycle of the process (blue curve) are reported. Both curves show that the total environmental benefit (reduction in the acidification indicator) increases at a decreasing rate compared to the increase of abatement costs when lowering the ELVs for HCl and SO₂, i.e., the marginal benefit of any additional euro spent in acid gas removal progressively decreases. This trend is remarkably more pronounced when the life cycle burdens are included in the analysis. **Figure 8** thus confirms the importance of including indirect life cycle burdens when determining the real cost-benefit relationship of emission policies, in an integrated pollution prevention and control framework.

As already evidenced in section 4.2, the situation across WtE plants is highly heterogeneous in terms of current emissions and type of acid gas removal technology, hence the cost-benefit considerations of a blanket policy of emission reduction equal for all the 36 Italian WtE plants, as the scenarios analysed in this study, could be different from plant to plant. **Figure 9** reports a marginal abatement cost curve, MACC (Kesicki and Strachan, 2011), calculated considering the different plants included in the case study. In the figure, the MACCs in each bar represents the contribution of a WtE plant to the net reduction of acidification impacts (life cycle perspective) compared to the baseline scenario. The height shows the additional abatement cost per $\text{kg}_{\text{SO}_2, \text{eq.}}$ removed in that plant to comply with the new emission scenario (S6 for Fig. 9a, S10 for Fig. 9b). The width of each bar is related to the reduction in the acidification impact in terms of $\text{kg}_{\text{SO}_2, \text{eq.}}/\text{y}$. Therefore, the area of the bar results as the total additional abatement cost for that plant compared to the baseline scenario. Wider bars indicate high potential to abate acid gas emissions. Shorter bars indicate high marginal abatement efficiency in terms of $\text{€/kg}_{\text{SO}_2, \text{eq.}}$.

The MACCs reported in **Figure 9** highlight that the majority of the additional costs associated with the two emission scenarios considered in the figure (S6 and S10) are generated by a limited number of plants. For example, in scenario S6 (Fig. 9a) more than 60% of the total costs are borne by 4 installations (P1, P6, P8, and P14; Table S12 in the SI reports the main features of the WtE facilities considered in the case study). The MACCs also provide indications on the costs and benefits of lowering ELVs for acid gases in single plants. For example, plants P9 and P18 provide a similar reduction of the acidification impact in scenario S6 (2 110 and 2 205 $\text{kg}_{\text{SO}_2, \text{eq.}}/\text{y}$, respectively), but the additional abatement cost for P18 is estimated to be approximately 20% of that of P9 (13.5 vs. 59.4 $\text{€/ kg}_{\text{SO}_2, \text{eq.}}$), due to the lower demand of reactants associated with a two-stage lime + bicarbonate treatment system (technology T4) compared to a single stage bicarbonate system (T3). In general, wet treatment systems (T6) and multi-stage dry (T4, T5) or dry + wet (T7, T8) technologies appear to require lower additional costs in the new emission scenarios. This does not imply that wet and multi-stage technologies are inherently better than dry technologies. In particular, wet

technologies are associated with higher capital costs, as evidenced by several studies (Dal Pozzo et al., 2018; Dong et al., 2020; Grieco and Poggio, 2009). However, in the context of the case study considered, focused on existing plants which are assumed to not incur in capital costs to comply with new emission scenarios, both wet technologies and multi-stage dry or dry + wet technologies can deal with lower ELVs with a limited over-consumption of reactants compared to single-stage dry systems.

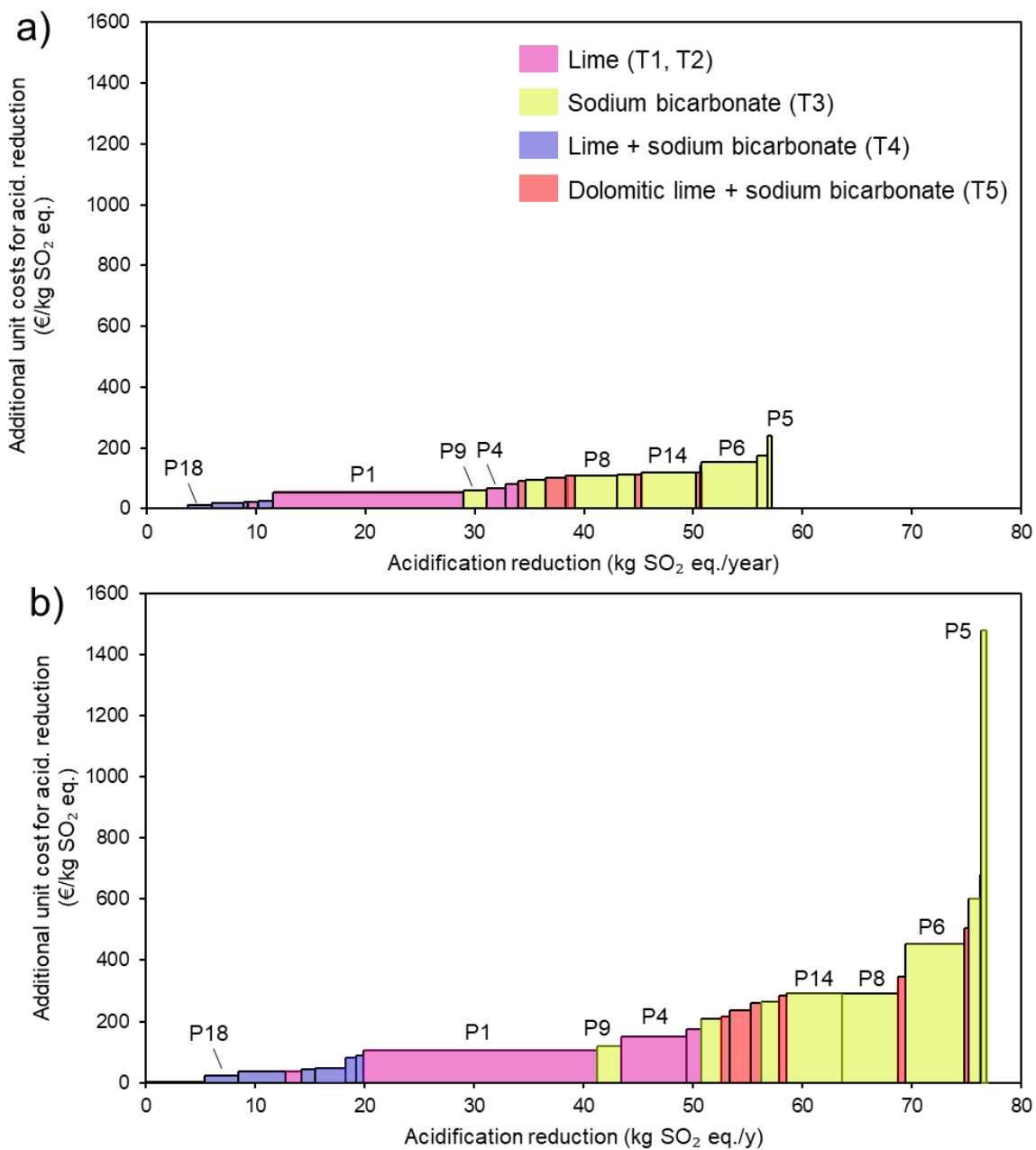


Figure 9. Marginal abatement cost curve (MACC) for acidification from S0 to S6 (a) and from S0 to S10 (b). Emission scenarios considered are described in **Table 3**. **Table S12** in the SI reports the more relevant data of each plant considered.

5. Conclusions

A methodology was developed for the assessment of cross-media effects associated to the introduction of lower emission standards for acid gas emissions in WtE plants. The combination of a simplified process modelling approach with a LCA-based characterization of the environmental burdens allowed tracking the variation of the overall impacts as a function of emission limit values at the stack of the plants. Applying the methodology to the whole Italian MSWI sector, it was possible to evidence the important role of the production chain of reactants needed for acid gas removal from flue gas in determining the overall impacts of the system. Technology-related patterns in the generation of cross-media effects were pinpointed and the costs and environmental benefits of the applications of new limits to the different plants were quantified.

The results obtained show that a further decrease of the acid gas ELVs of the Italian WtE plants, whose average HCl emission levels are often comparable with the lower end of the BAT-AEL range in the current emission scenario, would not result in a clear overall environmental benefit. Actually, when the stricter emission scenario considered in the case study is assessed, even if a 22% reduction in the acidification impact is obtained, a 20% increase in global warming and resource depletion impacts and a 21% increase in the operating costs of acid gas treatment are observed. The analysis clearly evidences the influence of country-specific variables in the determination of the results, hence the need to take into account local conditions. For instance, for countries with higher current acid gas emission levels than Italy or with a higher share of wet-based treatment systems, the overall advantage of lowering acid gas ELVs is expected to be clearer.

In general, the results of the analysis call for a thorough consideration of cross-media effects in the ongoing environmental permitting renewal process for existing WtE facilities started by the release of 2019 BREF WI. In this framework, the findings are of interest for environmental agencies, plant operators and technology suppliers alike. Environmental agencies may consider the trade-offs that each plant faces in the adaptation to new ELVs, based on its location, waste feed, choice of technology, and tailor the issuing of new plant-specific limits taking into account the full environmental implications. Plant operators should quantitatively assess the relationship between

economic costs and environmental benefits, in order to verify the convenience on both dimensions of alternative management options, e.g., on the destination of process residues. Technology suppliers should identify the emission levels that might be critical for a given technology or reactant and retrieve the necessary information to propose solutions at the WtE plant level (e.g., suggesting a retrofit to convert a single-stage treatment system in a multi-stage one) or at the life cycle level (e.g., focusing on the environmental hotspots of the supply chain).

Although the tools applied in the case study are specific for the assessment of cross-media effects generated by acid gases removal in WtE plants, the overall approach developed has a general validity and may be applied to similar problems (e.g., the regulation of acid gas emissions in other sectors, as the ceramics and glass industry, and/or the regulation of other pollutants in the WtE, as NO_x or mercury emissions), thus allowing the implementation of a holistic framework in the assessment of the actual environmental benefits deriving from the introduction of specific environmental measures.

References

- Ali, J., Rasheed, T., Afreen, M., Anwar, M.T., Nawaz, Z., Anwar, H., Rizwan, K., 2020. Modalities for conversion of waste to energy — Challenges and perspectives. *Sci. Total Environ.* 727, 138610.
- Ardolino, F., Boccia, C., Arena, U., 2020. Environmental performances of a modern waste-to-energy unit in the light of the 2019 BREF document. *Waste Manage.* 104, 94-103.
- Bacci di Capaci, R., Pannocchia, G., Dal Pozzo, A., Antonioni, G., Cozzani, V., 2022. Data-driven Models for Advanced Control of Acid Gas Treatment in Waste-to-energy Plants. *IFAC-PapersOnLine* 55(7), 869–874
- Beylot, A., Hocher, A., Michel, P., Descat, M., Ménard, Y., Villeneuve, J., 2018. Municipal Solid Waste Incineration in France: An Overview of Air Pollution Control Techniques, Emissions, and Energy Efficiency. *J. Ind. Ecol.* 22, 1016-1026.

- Biganzoli, L., Racanella, G., Rigamonti, L., Marras, R., Grosso, M., 2015. High temperature abatement of acid gases from waste incineration. Part I: Experimental tests in full scale plants. *Waste Manage.* 36, 98-105.
- Bisinella, V., Nedenskov, J., Riber, C., Hulgaard, T., Christensen, T., 2022. Environmental assessment of amending the Amager Bakke incineration plant in Copenhagen with carbon capture and storage. *Waste Manage. Res.* 40, 78-95.
- Bogush, A., Stegemann, J.A., Wood, I., Roy, A., 2015. Element composition and mineralogical characterisation of air pollution control residue from UK energy-from-waste facilities. *Waste Manage.* 36, 119-129.
- Bonfim-Rocha, L., Silva, A.B., Bernardo de Faria, S.H., Fernandes Vieira, M., de Souza, M., 2020. Production of Sodium Bicarbonate from CO₂ Reuse Processes: A Brief Review. *Int. J. Chem. Reactor Eng.*, 18, 20180318.
- Brivio, S., Kahale, N., Lombardi, L., 2018. Analisi del ciclo di vita di sistemi di rimozione dei gas acidi a secco in impianti di termovalorizzazione dei rifiuti urbani (in Italian). Venice 2018 Symposium, International Symposium on Energy from Biomass and Waste, 15-18 October 2018, conference proceedings.
- Cakir, N., Alp, E., Yetis, U., 2020. Assessing technologies for reducing dust emissions from sintermaking based on cross-media effects and economic analysis. *Clean Technol Environ Policy* 22, 1909-1928.
- Chen, C., Wen, Z., Wang, Y., Zhang, W., Zhang, T., 2022. Multi-objective optimization of technology solutions in municipal solid waste treatment system coupled with pollutants cross-media metabolism issues. *Sci. Total Environ.* 807, 150664.
- Chin, T., Yan, R., Liang, D.T., Tay, J.H., 2005. Hydrated Lime Reaction with HCl under Simulated Flue Gas Conditions. *Ind. Eng. Chem. Res.* 44, 3742-3748.

- Crenna, E., Secchi, M., Benini, L., Sala, S., 2019. Global environmental impacts: data sources and methodological choices for calculating normalization factors for LCA. *Int. J. LCA* 24, 1851-1877.
- Dal Pozzo, A., Antonioni, G., Guglielmi, D., Stramigioli, C., Cozzani, V., 2016. Comparison of alternative flue gas dry treatment technologies in waste-to-energy processes. *Waste Manage.* 51, 81-90.
- Dal Pozzo, A., Guglielmi, D., Antonioni, G., Tugnoli, A., 2017. Sustainability analysis of dry treatment technologies for acid gas removal in waste-to-energy plants. *J. Cleaner Prod.* 162, 1061-1074.
- Dal Pozzo, A., Guglielmi, D., Antonioni, G., Tugnoli, A., 2018. Environmental and economic performance assessment of alternative acid gas removal technologies for waste-to-energy plants, *Sustainable Prod. Consumption* 16, 202– 215.
- Dal Pozzo, A., Armutlulu, A., Rekhtina, M., Müller, C.R., Cozzani, V., 2018. CO₂ Uptake Potential of Ca-Based Air Pollution Control Residues over Repeated Carbonation–Calcination Cycles. *Energy Fuels* 32, 5386-5395.
- Dal Pozzo, A., Moricone, R., Tugnoli, A., Cozzani, V., 2019. Experimental Investigation of the Reactivity of Sodium Bicarbonate toward Hydrogen Chloride and Sulfur Dioxide at Low Temperatures. *Ind. Eng. Chem. Res.* 58, 6316-6324.
- Dal Pozzo, A., Lazazzara, L., Antonioni, G., Cozzani, V., 2020. Techno-economic performance of HCl and SO₂ removal in waste-to-energy plants by furnace direct sorbent injection. *J. Hazard. Mater.* 394, 122518.
- Dal Pozzo, A., Muratori, G., Antonioni, A., Cozzani, V., 2021. Economic and environmental benefits by improved process control strategies in HCl removal from waste-to-energy flue gas, *Waste Manage.* 125, 303-315.
- Dal Pozzo, A., Cozzani, V., 2021. Wastewater management of wet scrubbers in waste-to-energy facilities: a life cycle analysis. *Chem. Eng. Trans.* 86, 619-624.

- De Greef, J., Villani, K., Goethals, J., Van Belle, H., Van Caneghem, J., Vandecasteele, C., 2013. Optimising energy recovery and use of chemicals, resources and materials in modern waste-to-energy plants. *Waste Manage.* 33, 2416-2424.
- Dong, J., Jeswani, H.K., Nzihou, A., Azapagic, A., 2020. The environmental cost of recovering energy from municipal solid waste. *Applied Energy* 267, 114792.
- Dong, J., Tang, Y., Ruan, A., Chen, J., Lin, X., Lu, S., Gao, X., 2021. Environmental consequences of an ultra-low emission retrofit in coal-fired power plants from a life cycle perspective. *Waste Diposal Sustainable Energy* 3, 309-323.
- E-PRTR, 2021. European Pollutant Release and Transfer Register. Now hosted by the EU Industrial Emissions Portal. Available at: <https://industry.eea.europa.eu/> (Accessed: 13-05-2022).
- European Commission, Joint Research Centre, Cusano, G., Roudier, S., Neuwahl, F., et al., 2020. Best Available Techniques (BAT) reference document for waste incineration: Industrial Emissions Directive 2010/75/EU (Integrated Pollution Prevention and Control), Publications Office. Available at: <https://data.europa.eu/doi/10.2760/761437> (Accessed: 13-05-2022).
- European Commission, 2014. Life Cycle Data Network. Available at: <https://eplca.jrc.ec.europa.eu/LCDN/index.xhtml> (Accessed: 13-05-2022).
- Ferreira, F., Avelino, C., Bentes, I., Matos, C., Teixeira, C.A., 2017. Assessment strategies for municipal selective waste collection schemes. *Waste Manage.* 59, 3-13.
- Fruergaard, T., Hyks, J., Astrup, T., 2010. Life-cycle assessment of selected management options for air pollution control residues from waste incineration. *Sci. Total Environ.* 408, 4672-4680.
- Geldermann, J., Jahn, C., Spengler, T., Rentz, O., 1999. Proposal for an Integrated Approach for the Assessment of Cross-Media Aspects Relevant for the Determination of "Best Available Techniques" BAT in the European Union. *Int. J. LCA* 4, 94-106.

- Garcia-Herrero, I., Margallo, M., Onandia, R., Aldaco, R., Irabien, A., 2017. Environmental challenges of the chlor-alkali production: Seeking answers from a life cycle approach. *Sci. Total Environ.* 580, 147-157.
- Geldermann, J., Rentz, O., 2008. Multi-criteria Analysis for Technique Assessment: Case Study from Industrial Coating. *J. Ind. Ecology* 9, 127-142.
- Grieco, E., Poggio, A., 2009. Simulation of the influence of flue gas cleaning system on the energetic efficiency of a waste-to-energy plant. *Appl. Energy* 86, 1517-1523.
- Guinée, J.B. (Ed.), Gorée, M., Heijungs, R., Huppes, G., Kleijn, R., de Koning, A., Van Oers, L., Wegener Sleeswijk, A., Suh, S., Udo de Haes, H.A, De Bruijn, J.A., Van Duin R., Huijbregts, M.A.J. (2002). *Handbook on Life Cycle Assessment: Operational Guide to the ISO Standards. Series: Eco-efficiency in industry and science.* Kluwer Academic Publishers, Dordrecht (Netherlands).
- Horton, A.A., Walton, A., Spurgeon, D.J., Lahive, E., Svendsen, C., 2017. Microplastics in freshwater and terrestrial environments: Evaluating the current understanding to identify the knowledge gaps and future research priorities. *Sci. Total Environ.* 586, 127-141.
- IPCC, 2001. Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories. Technical report, Intergovernmental Panel on Climate Change. Available at: <https://www.ipcc-nggip.iges.or.jp/public/gp/english/> (Accessed: 06-03-2022).
- Kesicki, F., Strachan, N., 2011. Marginal abatement cost (MAC) curves: confronting theory and practice. *Environ. Sci. Policy* 14, 1195-1204.
- Liu, H., Li, S., Guo, G., Gong, L., Zhang, L., Qie, Y., Hu, H., Yao, H., 2022. Ash formation and the inherent heavy metal partitioning behavior in a 100 t/d hazardous waste incineration plant. *Sci. Total Environ.* 814, 151938.
- Luo, H., Cheng, Y., He, D., Yang, E.-H., 2019. Review of leaching behavior of municipal solid waste incineration (MSWI) ash. *Sci. Total. Environ.* 668, 90-103.

- Malinauskaite, J., Jouhara, H., 2019. The trilemma of waste-to-energy: A multi-purpose solution. *Energy Policy* 129, 636-645.
- Margallo, M., Massoli Taddei, M.B., Hernandez-Pellon, A., Aldaco, R., Irabien, A., 2015. Environmental sustainability assessment of the management of municipal solid waste incineration residues: a review of the current situation. *Clean Technol. Environ. Policy* 17, 1333-1353.
- Nedkvitne, E.N., Borgan, Ø., Eriksen, D.Ø., Rui, H., 2021. Variation in chemical composition of MSWI fly ash and dry scrubber residues. *Waste Manage.* 126, 623-631.
- Phungrassami, H., Usubharatana, P., 2021. Environmental Problem Shifting Analysis of Pollution Control Units in a Coal-Fired Powerplant Based on Multiple Regression and LCA Methodology. *Sustainability* 13, 5142.
- Pacher, C., Weber-Blaschke, G., Mocker, M., Quicker, P., 2009. Comparative ecological examination of different types of feedstock in dry sorbent injection processes for flue gas treatment (in German)- Final Report for the Federation of German Lime Industry, Technische Universität München-ATZ Entwicklungszentrum.
- Poggio, A., Grieco, E., 2010. Influence of flue gas cleaning system on the energetic efficiency and on the economic performance of a WTE plant. *Waste Manage.* 30, 1355-1361.
- Quicker, P., Rotheut, M., Noël, Y., Schulten, M., Athmann, U., 2014. Treating WTE flue gases with sodium bicarbonate. *Power Mag.* 158, 59–63.
- Quina, M.J., Garcia, R., Simões, A.S., Quinta-Ferreira, R.M., 2020. Life cycle assessment of lightweight aggregates produced with ashes from municipal solid waste incineration. *J. Mater. Cycles Waste Manage.* 22, 1922-1931.
- Rubin, E., McMichael, F.C., 1978. Cross-Media Environmental Impacts of Air Pollution Regulations for a Coal-Fired Power Plant. *J. Air Pollut. Control Assoc.* 28, 1099-1105.
- Sala, S., Cerutti, A.K., Pant, R., 2018. Development of a weighting approach for the Environmental Footprint. JRC Technical Report, Publications Office of the European Union, Luxembourg.

- Scipioni, A., Mazzi, A., Niero, M., Boatto, T., 2009. LCA to choose among alternative design solutions: The case study of a new Italian incineration line. *Waste Manage.* 29, 2462-2474.
- Ting, C.-H., Chen, H.-H., Yen, C.-C., 2008. A PID ratio control for removal of HCl/SO_x in flue gas from refuse municipal incinerators. *Control Eng. Practice* 16, 286-293.
- Van Caneghem, J., De Greef, J., Block, C., Vandecasteele, C., 2016. NO_x reduction in waste incinerators by selective catalytic reduction (SCR) instead of selective non catalytic reduction (SNCR) compared from a life cycle perspective: A case study. *J. Cleaner Prod.* 112, 4452-4460.
- Van Caneghem, J., Van Acker, K., De Greef, J., Wauters, G., Vandecasteele, C., 2019. Waste-to-energy is compatible and complementary with recycling in the circular economy. *Clean Technol. Environ. Policy* 21, 925-939.
- Vehlow, J., 2015. Air pollution control systems in WtE units: An overview. *Waste Manage.* 37, 58-74.
- Wang, D., Tang, Y.-T., Long, G., He, J., Robinson, D. (2020). Future improvements on performance of an EU landfill directive driven municipal solid waste management for a city in England. *Waste Manage.* 102, 452-463.
- Yuan, B., Hu, H., Huang, Y., Guo, G., Gong, L., Zou, C., Dong, L., Yao, H., 2022. Effect of hydrated lime on the migration behavior of selenium in the MSWI plants: Experimental and theoretical study. *Chem. Eng. J.* 433, 134603.