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Article

Techno-Economic Evaluation of Co-Removal of NO_x and SO_x Species from Flue Gases via Enhanced Oxidation of NO by ClO_2—Case Studies of Implementation at a Pulp and Paper Mill, Waste-to-Heat Plant and a Cruise Ship

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Abstract: Co-absorption of NO_2 and SO_2 from flue gases, in combination with the enhanced oxidation of NO by ClO_2(g), is studied for three different flue gas sources: a medium sized waste-to-heat plant; the kraft recovery boiler of a pulp and paper mill; and a cruise ship. Process modeling results are used to present the technical potential for each site together with cost estimation and optimization using a bottom-up approach. A process set-up is proposed for each site together with equipment sizing and resulting flows of process fluids. The simulation results, supported by experimental results, show that removal rates equal to or greater than current best available technologies are achievable with more than 90% of NO_x and 99% of SO_2 removed from the flue gas. The resulting cost of removing both NO_x and SO_2 from the flue gases is 2100 €/ton for the waste-to-heat plant, 800 €/ton for the cruise ship and 3900 €/ton for the recovery boiler. The cost estimation show that the consumption and cost of chemical additives will play a decisive role in the economic feasibility of the investigated concept, between 50% and 90% of the total cost per ton acid gas removed.

Keywords: flue gas cleaning; co-absorption; NO_x; SO_2; gas-phase oxidation; ClO_2

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1. Introduction

The relevance of control technologies for nitrogen and sulfur oxide (NO_x and SO_x) emissions remains high. Emission targets are continuously becoming increasingly strict. In the European Union, a national emission reduction commitment (NEC) directive is in force which specifies national emission reduction levels of NO_x and SO_x for each member state to comply with the 2020 and 2030 targets [1]. Currently 12 of the EU28 states need to decrease NO_x emissions to reach the 2020 goal and every state needs to decrease NO_x emissions to reach the 2030 goal, a majority of which need to reduce levels by more than 30% [2]. To reach the emission targets, reduction across every sector is required. The more stringent requirements on sources with current emission control systems employed or implementation of NO_x and SO_x control on point sources without previous emission control systems will arguably require new technologies suited for these applications.

Over the last decade, multipollutant emission control systems to reduce operating and capital costs of flue gas cleaning have been given much attention as part of a possible solution to the problem [3–6]. The idea is to remove NO_x and SO_x concomitantly in one unit. The combined removal of NO_x and SO_x is generally proposed to take place in a wet scrubber preceded by an oxidation step where NO is oxidized to NO_2. Several studies have shown that the co-absorption of SO_x and NO_x work for a variety of flue gas compositions and process conditions. Our research group studied the combined absorption of NO_x and SO_x from the flue gases of a waste incineration plant using gaseous chlorine dioxide (ClO_2) to oxidize NO to NO_2. It was shown that >90% of the SO_x and >80% of the NO_x can be removed using NaOH and Na_2SO_3 as scrubber additives [7]. Meng et al. studied the removal of NO_x and SO_x from a simulated flue gas using ozone (O_3) to oxidize NO. They used a steel slag slurry together with thiosulfate $S_2O_3{}^{2-}$ to increase the absorption and reached 100% of SO_x removal and >78% of NO_x removal [8]. In another study Hutson et al. investigated the simultaneous removal of SO_2, NO_x and Hg from a simulated coal flue gas using aqueous sodium chlorite ($NaClO_2$). They showed that complete removal of SO_2 and Hg can be achieved while removing about 60% of the NO_2 [9]. Furthermore, Si et al. performed experiments at pilot scale, 5000 Nm^3/h, using a newly designed scrubber where SO_2 and NO_x was removed using O_3 for oxidation of NO. They confirmed that increasing amount of SO_2 in the flue gas increases NO_2 absorption and they reached almost complete SO_2 absorption and >80% absorption of NO_x [10].

However, despite the amount of research and technical readiness, no techno-economic evaluation of the simultaneous removal of SO_x and NO_x has been published to the best of the authors' knowledge. Therefore, this work aims to conceptualize the design and derive cost estimations for end users. In this study, reaction modeling of the co-absorption of NO_x and SO_x is used to make techno-economic performance estimates of the proposed concept to evaluate economic feasibility for three different cases: a waste-to-heat plant; the kraft recovery boiler of a pulp and paper mill; and a cruise ship.

2. Methods

Techno-economic evaluation of simultaneous removal of NO_x and SO_x from flue gases were performed for three applications: a medium-sized waste-to-heat plant (20 megawatt, MW); a medium-sized pulp and paper mill (500,000 tons pulp/year) recovery boiler; and a cruise ship (12 MW). The technical performance of each case was investigated through simulations based on previous experimental results [7]. Economic evaluations were then performed based on the technical study. Each application had important differences in operating conditions and in emission standards and that is why the proposed process design, technological and economic performance varied between cases.

2.1. Case Studies

2.1.1. Waste-to-Heat Plant

The waste-to-heat plant operates with an inhomogeneous fuel inherently creating variations in flue gas composition making NO_x control more challenging. In Sweden, there is a charge on NO_x of 4900 €/ton of NO_x emitted together with an abatement system which returns 0.83 €/MWh (useful) to the emitter with the consequence that there is a cost for emitting more than 0.17 g NO_x/kWh. If NO_x can be prevented at a cost <4900 €/ton, profit can be made. The current best available technology (BAT) for NO_x removal is selective catalytic reduction (SCR) with removal rates of ~90% and selective non catalytic reduction (SNCR) with removal rates of <50% [11]. For SO_2 removal, BAT consists of 5 different technologies, including wet flue gas desulfurization (WFGD) with removal of >99% [11]. Both NO_x and SO_x removal technologies are to be regarded as available and technically feasible.

2.1.2. Kraft Recovery Boiler

The emissions from the kraft recovery boiler are regulated in the permit to operate where a maximum allowed limit of kg NO_2 emitted per produced ton of pulp produced is set. The limit is set by the local authorities and varies depending on age and location of plant. There are ongoing investigations on enforcing national levels, which will require exhaust gas treatment systems. Increasing emission standards can also be seen globally, where some pulp and paper mills in China are being required to install NO_x control on kraft recovery boilers [12]. Kraft recovery boilers have low SO_2 emissions which is not favorable for the proposed technology, but they have processes and facilities in place and long experience of handling the proposed process chemicals. There is also the opportunity for waste treatment in the plants' existing sulfur recirculation, which makes the proposed technology more interesting from a business case perspective. The BAT for SO_2 control from kraft recovery boilers is WFGD with recirculation of the sulfur, achieving >90% removal of SO_2 [13]. Current BAT for NO_x control is SNCR with >50% removal of NO_x. SCR is discussed as a high removal rate alternative (>80% removal) but without current implementations in Europe [13]. The high dust content in the recovery boiler provides a difficult environment for SCR operation and requires additional dust removal units.

2.1.3. Cruiser

The cruiser differs from the two other cases in that the requirements will differ depending on geographic positioning. The international maritime organization (IMO) has standards that are detailed in annex VI for both SO_2 and NO_x emissions from shipping detailed in Regulations 14 and 13, respectively [14,15]. In addition to the agreed global limits that are binding for ratifying states, specific emission control areas (ECAs) also exist where the emission limits are considerably lower compared to the global standards. In annex VI, regulation 14 the SO_2 emissions are to either be controlled through the use of low sulfur-containing fuels or by the usage of exhaust gas treatment systems, generally scrubbers. The emission limits are set equivalent to a fuel sulfur content of 0.1% m/m in ECAs and 0.5% m/m globally. In the IMO annex VI, regulation 13 the NO_x emission standards are divided into three tiers related to engine speed, where tiers 2 and 3 are currently in place and tier 1 is outdated. Tier 2 (rpm >2000) is the globally enforced limit at 7.7 g NO_x/kWh and tier 3 (rpm >2000), which limits emissions of NO_x to 2 g/kWh is only enforced in ECAs. From an economic perspective this makes a flexible exhaust gas treatment system attractive since the stricter emission limits can be met when required and expenses can be spared outside of ECAs. The current BAT for NO_x control on cruisers is SCR with current installations in operation.

2.2. Choice of Oxidizing Agent

It is vital for the absorption of NO_x that NO is oxidized to NO_2 to increase its solubility. Different chemicals have been tested for NO oxidation, including ClO_2, H_2O_2 and O_3 [8,16,17]. The process integration of the oxidation step varies depending on the chemical used. For gas phase oxidation, H_2O_2 requires high temperatures, above 300 °C, for the oxidation to take place [18]. ClO_2 and O_3 can operate at lower temperatures, 70–160 °C while maintaining efficiency [16,19]. H_2O_2 and O_3 have an advantage compared to ClO_2 in that preferable reaction products are formed, H_2O and O_2, compared to the HCl formed from ClO_2, but selectivity and reactivity with respect to NO to NO_2 oxidation is still below that of ClO_2 [20]. The high oxidation potential of O_3 has been shown to oxidize NO to higher oxidation states beyond NO_2 if the added O_3 exceeds a molar ratio with NO of 1.2 [21].

In this study ClO_2 was used in the oxidation step due to the previous research on ClO_2 by our group that shows high efficiency in a lower temperature range and simple process integration of the oxidation [7]. However, it should be noted that all aforementioned oxidation chemicals appear to be viable options and depending on regional legislations, site facilities and flue gas temperatures, one oxidizing agent might be favored above the others. In Table 1 the three discussed oxidizing agents are compared based on efficiency. While both $NaClO_2$(aq) and H_2O_2(aq) are commonly used in experiments only gaseous oxidation is included since the consumption, translating into efficiency, of aqueous oxidizing agents is seldom reported in the literature. Often concentration is used to determine the efficiency of NO oxidation but without specifying the amount of oxidizing agent continuously added to maintain this concentration.

Table 1. List of oxidizing agents to be considered and associated technical results. OA stands for oxidizing agent and the "OA/NO" is the molar ratio between OA and NO in the flue gas.

Method	NO Oxidation Rate	Main Active Species	Oxidizing Agent Efficiency (OA/NO)	Reference
ClO_2	99%	ClO, ClO_2	0.5	[7]
O_3	95%	O_3	1	[8]
O_3	95%	O_3	1.25	[22]
H_2O_2	95%	H_2O_2	2.7	[17]

2.3. Process Designs and Contingencies

Figure 1 (upper) shows a conceptual design of a combined NO_x and SO_x control system for a waste-to-heat plant and a recovery boiler where ClO_2 is used to enhance NO to NO_2 oxidation. ClO_2 is injected to the main flue gas path, preferably at a point where the flue gas temperature is 150 ± 10 °C in order to maximize efficiency and minimize the risk of corrosion. In the conceptual design the oxidation reactor is constructed of Stainless steel 304 L. The reactor should be designed to increase mixing with minimal pressure drop. It is noteworthy that results obtained in previous studies indicate that the oxidation of NO to NO_2 via ClO_2 is rapid and not dependent on an advanced mixing reactor design [16,23]. The flue gas continues to a quench where temperature decrease, and the gas becomes saturated. The quench is maintained at an acidic pH of 1.5 and is of sufficient height to absorb the vast majority of chloride present in the flue gas. A residence time of 5 s is used in this study for design of equipment. The quench is constructed using Stainless steel 6MO/SMO to withstand the harsh environment. The scrubber is designed to achieve 90% absorption of incoming NO_2. Sodium sulfite (Na_2SO_3) and sodium hydroxide (NaOH) is supplied directly to the liquid feed and not into the scrubber tank in order to minimize reactions with the liquid bulk. In this design, the scrubber is constructed of stainless steel SS2205 to ensure the lifetime of the equipment. The quench and scrubber tank is given dimensions such that it has a volume equal to 3 min of liquid flow [24]. Both tanks are constructed of SS2205. The chemical storage units for NaOH and Na_2SO_3 are designed to hold a volume equal to 1 week of operation. All pumps are centrifugal and sized to fit.

In the case of the cruise ship, the conceptual design is different and seen in Figure 1 (lower). SO_x emission control on ships usually allows both for closed and open loop operation, where the wastewater is stored for treatment while in closed loop and rejected to the surrounding water in open configuration. While the release of wastewater is allowed outside of ECAs, the system will need to be able to operate in such a way to be economically feasible. It is also desirable for the system to be able to run without scrubber operation so the system must allow for high temperatures. A quench is not deemed necessary to separate chloride since the cruiser uses sea water as scrubber solution.

For a complete list of equipment and sizing for each case see the Supplementary Materials. The sizing and installation of the chlorine dioxide production unit is not included in the study.

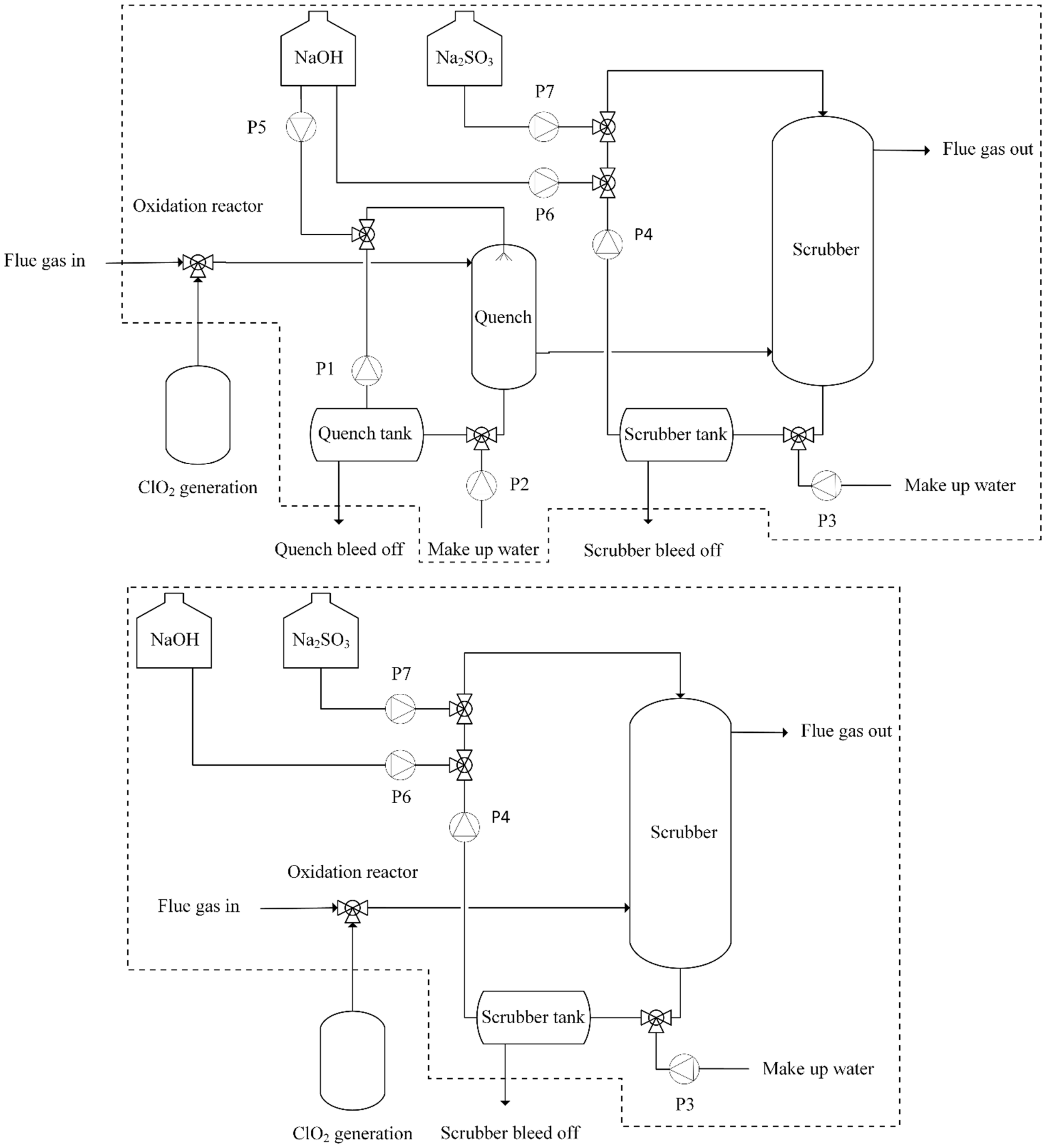

Figure 1. Principal process diagram of the implementation of co-removal of NO_x and SO_x from flue gases via enhanced oxidation of NO by ClO_2. The upper scheme illustrating the design at the waste-to-heat plant and the pulp and paper mill. The lower design illustrates that of a cruise ship where no quench is present. Dashed lines indicate system boundary for economic analysis where the cost associated to wastewater treatment is not included, additionally only the production cost of ClO_2 and not the production unit is included. For a complete list of sizing and materials, see Supplementary Materials.

2.4. Process Modeling

For each case, the sizing of the equipment and evaluation of the performance of the gas treatment process, the simulation software Aspen Plus V.11 was used. The simulations are based on the design criteria and flue gas concentrations available in Table 2. The removal efficiencies of SO_2 and NO_2 are set to >99% and >90%, respectively. The target of >90%

NO_2 removal is set as to match the performance of a SCR system and the SO_2 removal target is set close to 100% since this has been shown experimentally to follow with the NO_2 absorption. The flue gas concentrations used in the simulations are based on typical values for each application.

Table 2. Comparison of the parameters applied in the three different applications. HCl concentration corresponding to the amount of ClO_2 needed for NO to NO_2 oxidation.

Parameter	Waste to Heat Plant	Kraft Recovery Boiler	Cruise Ship
SO_2 removal efficiency (%)	>99	>99	>99
NO_2 removal efficiency (%)	>90	>90	>90
Raw Flue Gas Flow (Nm^3/h)	57,500	357,000	61,600
	Flue gas concentrations after oxidation		
N_2 (%)	65	68	76
CO_2 (%)	8	12	3.2
H_2O (%)	16	16	9.2
O_2 (%)	11	3	11.3
SO_2 (ppm wet)	150	20	900
NO_2 (ppm wet)	150	150	600
HCl (ppm wet)	60	60	220

The process model considers the reactions in the tanks, in addition to the mass transfer and reaction kinetics in the scrubber and quench. The quench is modeled as a packed tower with a high void fraction of 0.96 to minimize pressure drop. Both the quench and the scrubber tower have diameters designed based on an 80% approach to flooding to maximize the efficiency. The chemical reactions and eventual steady state composition of the bleed stream is not considered in this work. Each case is modeled.

The chemistry in the scrubber and quench, except for one reaction, is based on a previously developed mechanism and is discussed in detail elsewhere [25]. The reaction included in this analysis, and not in our prior work, is the oxidation of S(IV) to S(VI) by O_2. The oxidation of S(IV) to S(VI) in a combined NO_2–SO_2 removal system is discussed thoroughly in literature but the rate of the reaction is still debated. In our previous experiments we have seen that the oxidation of S(IV) is rapid without any addition of other chemicals. With addition of $Na_2S_2O_3$, the oxidation rate of sulfite is decreased. $Na_2S_2O_3$ is also an effective chemical for NO_2 absorption and we have in our experiments reached 90% NO_2 removal with $Na_2S_2O_3$ addition to the scrubber. However, there are no kinetics for a $Na_2S_2O_3/Na_2SO_3$ system for NO_2 absorption why only Na_2SO_3 is used in these simulations. To better match the rapid oxidation of S(IV) that has been observed, a worst-case scenario has been adopted when running the simulations. The reactions in which bisulfite (R1) and sulfite (R2) react with dissolved oxygen to form bisulfate and sulfate respectively were added to the scrubber chemistry as first order reactions in regards to S(IV) and O_2 with an activation energy of 29 kJ/mol and rate constant of 6E6 s^{-1}, which is the highest reported reaction rate that the authors have found in literature [26]. The reactions are defined as follows,

$$HSO_3^- + 0.5O_2 \rightarrow HSO_4^- \tag{R1}$$

and

$$SO_3^{2-} + 0.5O_2 \rightarrow SO_4^{2-}. \tag{R2}$$

The added reaction will have a significant impact on the overall chemistry and increase the amount of S(IV) needed to reach the NO_2 absorption criteria. With the added reaction, results are more in line with what has been observed during experiments using Na_2SO_3. However, it is assumed that all, or major parts of the Na_2SO_3 mentioned in this work will be substituted to $Na_2S_2O_3$ in future full-scale applications. The added amount of absorption reagent (Na_2SO_3 and $Na_2S_2O_3$) should, therefore, be expected to drop compared to the results presented here.

2.5. Cost Estimation

The cost of the acid gas (NO_2, SO_2) removal system, as shown in Figure 1 (dashed lines) is estimated using Aspen Process Economic Analyzer V11 based on the process model described using typical values [27]. The cost basis is for a nth of a kind plant and before taxation. Total direct cost (TDC) is the equipment cost together with piping, instrumentation, electrical etc., together with process contingencies. Total indirect cost includes construction and engineering together with project contingencies. The total direct cost together with indirect cost and owner's cost is equal to the total plant cost (TPC). The main assumptions in the economic analysis are presented in Table 3. In addition to TDC and TPC, total specific cost, which consists of annualized TDC plus variable operating cost, is defined to evaluate the cost of every ton of removed NO_2 and SO_2.

Cost of equipment is largely dependent on material choice. For the specification of the reviewed process, high grade materials have been used due to the corrosive nature of the solutions. The material and component specification for each case study is available as Supplementary Materials. Fixed operating and maintenance (O&M) cost, including all labor and maintenance, are estimated as percentages of TPC with estimates based on the values in Rao et al. [28]. Variable operation cost, including chemical reagents, water and electricity price is estimated based on available literature but might be subject to large deviation depending on local availability and scale of operation.

It should be noted that all assumptions are made for land-based installation and production. The cost variation based on marine application is not taken into account. The cruiser is not bound to be produced in the EU or to operate in the EU and, therefore, costs might change on the production and installation. The harsh environment at sea can increase the maintenance cost of the unit and operating hours will vary depending on the designated route. The chemical cost should remain similar as for land based installation and while electricity generation at sea will be based on the usage of fuel, prices per kWh are similar [29]. In addition to the uncertainties of the values in Table 3, the limited space available on cruisers will pose a challenge for storage/production of chemicals to be used in the process.

Table 3. Main assumptions for the economic analysis.

Project Region Base	EU
Project currency	€ (2020)
Conversion rate €/US. $	0.85
Lifetime of Unit	25 years
Tax rate	0%
Inflation rate	0%
Annual operating hours	8000
Process contingencies	10% [27]
Construction time	1 year
Indirect costs	14% of TDC [30]
Project contingencies	15% of TDC [27]
Owner's cost	7% of TDC [27]
Number of full-time employees required	0.5
Total Maintenance cost	2.5% of TPC [28]
NaOH 100 wt%	400 €/ton [1]
Na_2SO_3 100 wt%	500 €/ton [2]
ClO_2 100%	1200 €/ton [31]
Electricity	68.4 €/MWh
Make-up water	6.65 €/m^3 [30]

[1] Estimated average. Values range between ~200–600 €/ton. [2] Estimated average. Values range between ~400–1000 €/ton.

3. Results and Discussion

This section is presented in two parts, technical performance, and economic performance. In the technical performance, process design and absorption rates are investigated. The economic performance presents capital and operating cost for each application. However, it should be noted that the design is derived from in an iterative procedure between process and economic performance.

3.1. Technical Performance

The design of the proposed units of each case is based on the criteria in Table 2 and the design specification is available in Supplementary Materials. In Table 4 the resulting flows of water, liquid bleed concentration and supporting chemicals to reach 90% NO_x absorption, with scrubber dimensions specified in Supplementary Materials, are displayed.

The flows for each case are directly proportional to the size of the unit and the ratio and amount of SO_x and NO_x. The scrubber liquid flow is adjusted to maintain a constant liquid to gas ratio (L/G) at 10. The NaOH flow corresponds to the amount of acid gases absorbed and the Na_2SO_3 flow corresponds to the $SO_{x-}NO_x$ ratio in the flue gas. The relation between the variables is evident with the highest NaOH flow and lowest Na_2SO_3 flow for the cruiser, which has the highest SO_x-NO_x ratio and highest total amount of acid gases removed. The amount of ClO_2 is fixed at 0.6 kg ClO_2 per kg NO_2 in the flue gas.

The scrubber bleed concentration profile for the waste-to-heat plant and the pulp and paper mill is similar due to the similarities in flue gas concentrations. The two major components present are S(VI) and N(III) followed by hydroxylamine disulfonic acid (HADS) and carbonate. Only a minor amount of S(IV) is present and almost no N(V). The S(IV)/NO_2 molar ratio, defined as the total amount of moles sulfur added to the scrubber in relation to the moles of NO_2 in gas phase entering the scrubber, is higher for the waste to heat plant compared to the recovery boiler. However, the resulting liquid concentration of S(IV) is lower for the waste to heat plant. This is due to the higher O_2 concentration in the flue gas for the waste to heat plant and the increased importance of the oxidation of S(IV) to S(VI) via O_2 compared to the pulp and paper mill. For the cruiser in closed mode, the share of nitrogen components to sulfur components is slightly higher. With open mode, all concentrations are low compared to the closed mode, except for the concentration of S(IV) which is almost as high as for the waste-to-heat plant.

Table 4. Resulting scrubber flows and species concentration in scrubber bleed to reach design criteria for each application.

Parameter	Waste to Heat Plant	Kraft Recovery Boiler	Cruise Ship, Closed/Open
Liquid Flow (m^3/h)	575	3570	616
L/G (kg liquid/kg gas)	10	10	10
NaOH (l/h)	58	75	350/280
Na_2SO_3 (l/h)	60	460	10/150
S(IV)/NO_2 ratio [1]	1.3	1.2	1.7/2.2
ClO_2 (kg/h)	8.5	52.7	33.7/33.7
SO_2 removed (kg/h)	20	20	130/130
NO_2 removed (kg/h)	13	80	50/50
Make up water (m^3/h)	1.34	4.16	6.26/616
Liquid bleed (m^3/h)	1.38	6.1	6.27/616
N(III) (g/L)	8.8	10.5	7.9/0.1
N(V) (mg/L)	36	9.4	1.98/0.67
S(IV) (mg/L)	60	140	1110/90
S(VI) (g/L)	25	25	25/0.11
HADS (g/L)	2.7	10	3.4/0.0
Carbonates (g/L)	1.8	2.2	0.67/0.0

[1] S(IV)/NO_2 ratio is defined as the total amount of moles of sulfur added to the scrubber via Na_2SO_3 and SO_2 divided by the total amount of moles NO_2 in the gas phase entering the scrubber.

Figure 2 shows how the impact of scrubber pH on the liquid composition in the scrubber bleed. These simulations are for the waste-to-heat plant with operating conditions according to Table 2. The model predicts a maximal NO_2 absorption at a pH of 7.2. No differences are seen in SO_2 absorption in the pH-range from 6 to 8. The oxidation of absorbed SO_2 to S(VI) greatly increase the absorption capacity of SO_2 which is why the removal is always close to 100%. The increase in pH above 6 result in an exponentially increased amount of absorbed CO_2. Absorbed CO_2 will influence process economics negatively since the amount of NaOH added changes in relation to this. The model predicts an exponential decrease of HADS when increasing pH from 6 to 8. HADS is formed from HSO_3- and increased pH will shift the S(IV) equilibrium towards SO_3^{2-}. Sulfite is never present in the bottom liquid at concentrations above 150 mg/L and the same is true for N(V) which requires a more acidic solution to be present. The increase in N(III) and S(VI) with increasing pH corresponds to the decreased concentration of HADS.

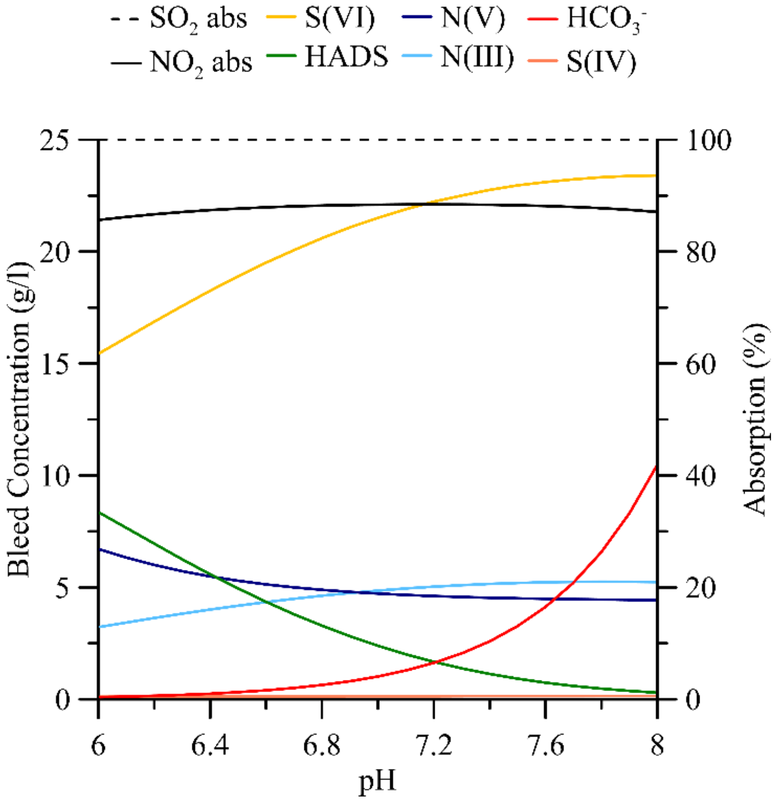

Figure 2. Liquid composition variations based on pH at scrubber bleed and resulting NO_2 and SO_2 absorption. Simulations are based on the waste-to-heat plant case.

Figure 3 shows how the NO_2 absorption will increase with increasing total amount of S(IV) added to the system either from Na_2SO_3 or SO_2 in the gas phase for the three cases. The molar ratio of added Na_2SO_3 to NO_2 is constant at 0.3 for the waste-to-heat plant (WTHP in the figure), 1.1 for the recovery boiler and 0.04 for the cruiser. NaOH addition is adjusted to maintain the pH at 7.2 for each simulation. The results indicate that the required addition of S(IV)/NO_2 (SO_2 and Na_2SO_3) is between 1.1 and 1.3 to reach 90% absorption of NO_2. The operating point for each case is indicated with a circle on the respective line. The waste-to-heat plant is the most sensitive to changes up or down around the assumed SO_2 concentration (operating position) with respect to NO_2 absorption and will need to adjust Na_2SO_3 addition to maintain constant absorption of NO_2. The operating conditions and design for the recovery boiler and the cruiser is relatively insensitive to changes in SO_2 concentration. The recovery boiler relies almost solely on Na_2SO_3 for NO_2 absorption and the amount is set at a level which reaches 90% NO_2 absorption. For the cruiser, the sulfur content in the cruiser fuel could be decreased with 25% and still be enough for 90% NO_2 absorption. Below a $SO_2\_NO_2$ ratio of 1.1, the NO_2 absorption drops rapidly for the cruiser.

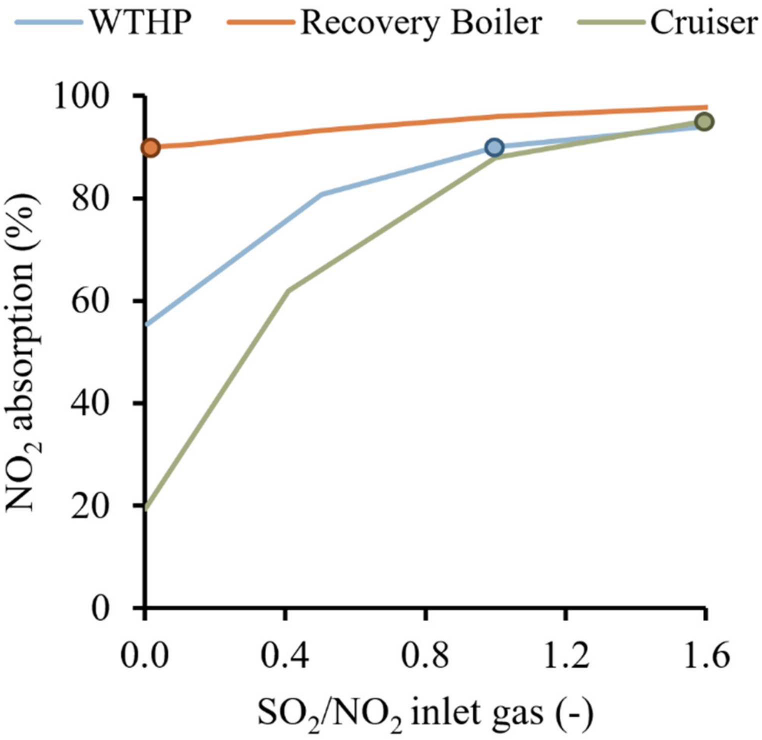

Figure 3. NO_2 absorption in relation to SO_2/NO_2 ratio in the flue gas. The NO_2 absorption is displayed for the three different cases as described in Table 2. The Na_2SO_3 addition is kept constant for each case at 0.3 for the waste-to-heat plant (WTHP in figure), 1.1 for the recovery boiler and 0.04 for the cruiser. Each circle shows the operating position for each respective case. NaOH dosing is adjusted to maintain constant pH at 7.2.

Figure 4 shows how the NO_x absorption will increase with increasing scrubber height for the three different case designs. The corresponding gas residence time in the scrubber changes linearly between 2 and 30 m of height from 1 to 17 s for the waste-to-heat plant and the recovery boiler and from 1 to 12 s for the cruiser. Gas composition, flow and added Na_2SO_3 remains constant while NaOH is changed to maintain pH constant at ~7.2. The increased absorption per meter added scrubber height is decreasing with each meter added, due to decreasing NO_2 partial pressure and S(IV) concentration. From the figure, the height of the scrubber for each final design case can be seen where the NO_2 absorption reaches 90% (except for the cruiser in closed mode which has 95% absorption). The difference between each case is due to the difference in S(IV)/NO_2 ratio in the scrubber where a higher S(IV)/NO_2 ratio will increase the reaction rate of NO_2 absorption. The cruiser has the highest S(IV)/NO_2 ratio and, therefore, requires the lowest height of the scrubber. The waste-to-heat plant design has the lowest S(IV)/NO_2 ratio and requires a much taller scrubber. Each S(IV)/NO_2 ratio is visible in Table 4.

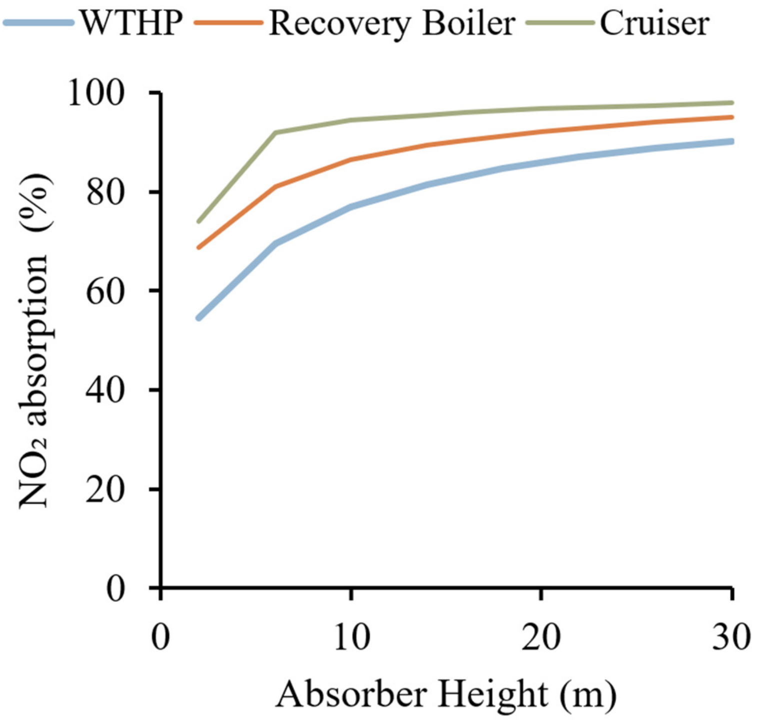

Figure 4. NO_x absorption in relation to scrubber height with all parameters retained according to Table 4. except for NaOH dosing to maintain pH constant at 7.2. Waste-to-heat plant abbreviated to "WTHP" in figure legend.

3.2. Economic Performance

Figure 5 shows the total specific cost of acid gas removal (combined SO_2 and NO_2) in €/(ton SO_x and NO_x) removed for all the investigated cases. The results are based on simulations that reach the design criteria of 90% NO_x removal and >99% SO_2 removal. It is apparent that chemical cost is a major factor when estimating the cost of the reviewed system. Approximately 50% of the total specific cost is related to the chemicals. For the waste-to-heat plant, the estimations give a cost of about 2100 €/ton acid gases. The cruiser has a cost of about 800 €/ton acid gases in closed mode with a slightly higher cost of about 900 €/ton acid gases for the open mode. In open mode, the NO_2 absorption efficiency per added kilogram of absorption agent is decreased compared to closed mode due to small amounts of remaining S(IV) being rejected. The recovery boiler case is the most expensive due to the low concentrations of SO_2 in the flue gas at about 3900 €/ton acid gases. Higher concentrations of SO_2 in the flue gas decrease this result in two ways. Firstly, the removal of SO_2 is cheaper, almost only increasing NaOH consumption and secondly it lowers the amount of Na_2SO_3 needed for NO_2 absorption. There is an opportunity to lower chemical cost for the recovery boiler through the usage of white liquor as an absorption reagent. The availability on site could prove cost efficient. However, it will not be possible to use with the design as indicated in Figure 1 since the high pH of white liquor (>12) will result in a substantial absorption of CO_2, as indicated by Figure 2. In a recent study, Hruška et al. investigated the usage of white liquor and NaOH to absorb H_2S from black liquor gasification [32]. They concluded that the amount of CO_2 absorbed by white liquor when used as an absorbent will increase the lime kiln load to a degree that will have significant negative impact on process economy.

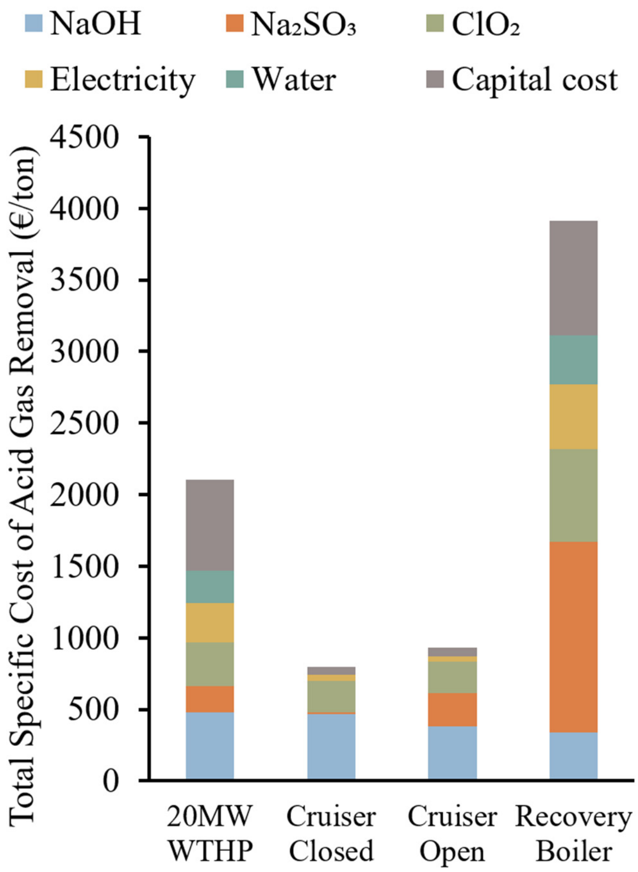

Figure 5. Total specific cost of acid gas removal in €/ton for all the investigated cases as described in Table 2 divided by expense category. Capital cost is the annualized TPC.

Figure 6 shows the total specific cost of NO_x removal in in €/ton removed. The result differs from that in Figure 5 by indirectly showing the amount of SO_2 that is removed by the process. The recovery boiler has the lowest SO_2 content in the gas while the cruise ship has the highest, shifting the total specific cost of NO_2 removal to increase to the highest level for the cruise ship and the lowest for the recovery boiler. The waste-to-heat plant has an estimated cost of ~5400 €/ton, the cruise ship has ~2700 and 3300 €/ton in closed and open modes, respectively, and the recovery boiler has an estimated cost at ~4700 €/ton.

When relating the result to the aforementioned NO_x charge in Sweden of 4900 €/ton, the result indicates economic viability for installation of co-removal of NO_x and SO_x at a Swedish waste-to-heat plant. The total specific cost of NO_x removal of 5300 €/ton would net a 400 €/ton loss when only NO_2 removal is considered. When also accounting for the SO_2 removal the relative cost looks competitive. In the waste-to-heat plant case, for each ton of NO_2 removed, 1.54 ton of SO_2 is also removed resulting in a cost of about 260 €/ton SO_2. In addition to this, a special case for the waste-to-heat plant is that the fuel is a source of income and lower-grade waste gives a higher income. This creates an incentive for being able to take in high sulfur containing wastes which will reduce the need for added Na_2SO_3.

The estimated specific cost of NO_2 removal for a recovery boiler unit is about 4700 €/ton which nets a 200 €/ton NO_2 profit, not even accounting for the SO_2 removal if the recovery boiler were to be included in the Swedish charge on NOx emission system. This application is the most sensitive to absorption reagent cost and effectiveness. Almost entirely relying on added sulfite as a source of active sulfur for NO_2 absorption the total specific cost of NO_x removal would decrease with around 20% if the added sulfur was reduced by 50%, something that might be possible with the addition of $Na_2S_2O_3$.

The target of 90% removal of NO_2 at the recovery boiler might be higher than what is required at certain sites. If the required NO_2 removal is lower than 90%, the cost of

Na_2SO_3 will drop significantly. However, if the required NO_2 removal is lower than 50%, selective non-catalytic reduction will almost certainly be a cheaper alternative, even with the challenging implementation.

The cruiser has the lowest estimated total specific cost of NO_2 removal due to the high amount of SO_2 present in the flue gas and the relatively cold scrubber liquid (sea water). The cost might also be further decreased in retrofitting cases since it is assumed that the scrubber to be used is the same design as currently installed SO_2 scrubbers. However, the economic feasibility for the ship application needs to be compared to other maritime alternatives. It is technically possible to reach the emission standards put in place by the IMO, but other techniques might be more affordable. The liquid treatment and storage when operating in closed mode should be regarded as an uncertainty in terms of cost.

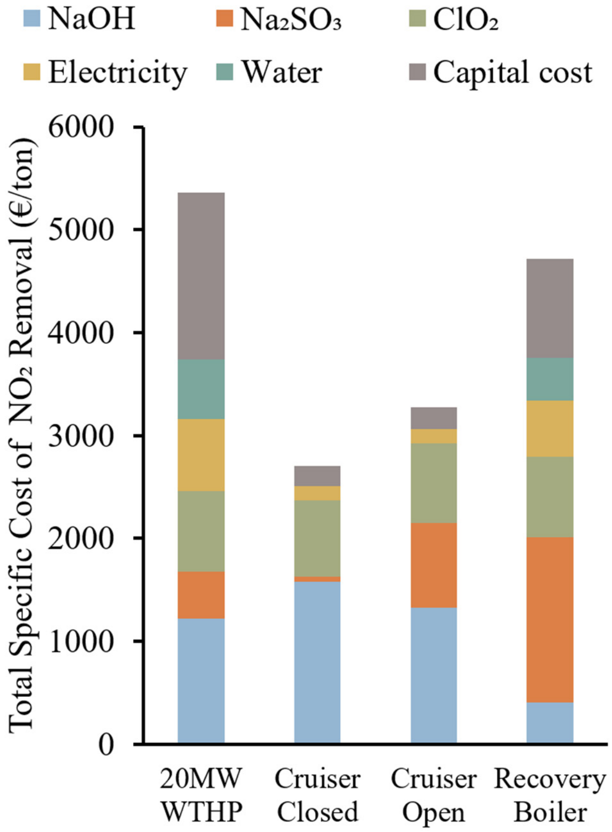

Figure 6. Total specific cost of NO_2 removal in €/ton for all the investigated cases as described in Table 2 divided by expense category. Capital cost is the annualized TPC.

Figure 7 shows the estimated total direct cost for the three different cases. The main cost is related to the scrubber with over 60% of the total cost in all applications. The quench, when present, is estimated at about 25% of the total cost and tanks and pumps equates to roughly 15% of the total cost. The investigated process optimized for the currently employed design and cost assumptions have a large share of the total cost related to capital cost. If the consumption of sulfite can be decreased compared to what is simulated and/or if the cost of sulfite is lower than what is assumed in this work then a lower scrubber height than what has been used in this work will be a more cost-efficient option, significantly lowering the capital cost of the process. A summary of cost results, including TDC, TPC, variable and fixed cost can be found in Supplementary Materials.

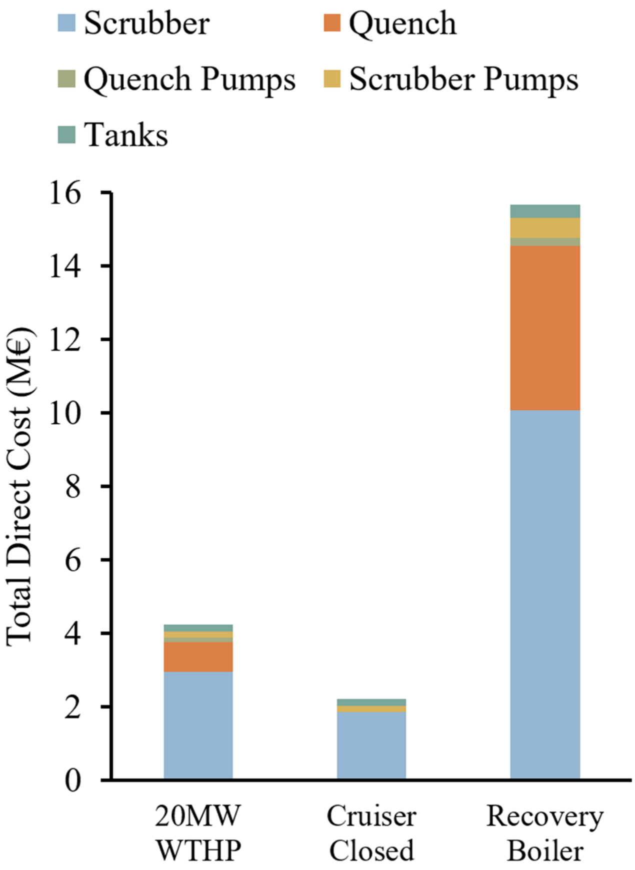

Figure 7. Total direct cost in M€ divided between equipment category for all the investigated cases as described in Table 2. The recovery boiler has substantially higher total direct cost compared to the other two cases due to the much larger size of the plant, about six times larger.

Figure 8 visualize the impact on removal cost by increasing the scrubber height from 6 to 40 m while maintaining 90% NO_2 and 100% SO_2 removal for the waste-to-heat plant design. At a scrubber height lower than 6 m it is impossible to maintain NO_2 absorption above 90% while at the same time having a discharge concentration of sulfate at 25 g/L since a major part of the sulfur will leave the scrubber as $SO_3{}^{2-}$. Therefore, scrubber heights below 6 m are excluded from the figure. For all other cases, concentrations and efficiencies are constant. The cost for NaOH and ClO_2 remains almost unchanged with scrubber height since the total amount of acid gases absorbed remains constant. At the same time, water and Na_2SO_3 cost decrease with increased scrubber height since less Na_2SO_3 is needed to be added and capital cost increase with added height. The total specific cost of NO_2 and combined SO_2/NO_2 removal is lowest, 5300 and 2100 €/ton respectively, at a scrubber height of 30–34 with the design conditions according to Table 2, "waste-to-heat plant". NaOH, includes the NaOH consumption for both the scrubber and the quench.

The absence of other techno-economic studies on the combined removal of NO_x and SO_x makes comparisons difficult. However, a comparison can still be made to other economic evaluations performed for only NO_x or SO_x removal. In 2017, Yaramenka et al. published a report in which they estimated the cost benefit of implementing SCR to cruise ships in the Baltic sea [33]. They estimated that a SCR system for NO_x control with 90% reduction would operate at a total specific cost ranging from 2000 to 5500 €/ton NO_x depending on the lifetime of the equipment, from 5 to 15 years, and interest rates. Figure 6 shows the corresponding values found in this study to 2700 and 3300 €/ton NO_x for closed and open scrubber configurations, respectively. The cost estimates for NO_x control on a cruiser in this study is close to those estimated for a SCR system with the same performance and operating conditions. However, the cost in this study also includes the removal of SO_2.

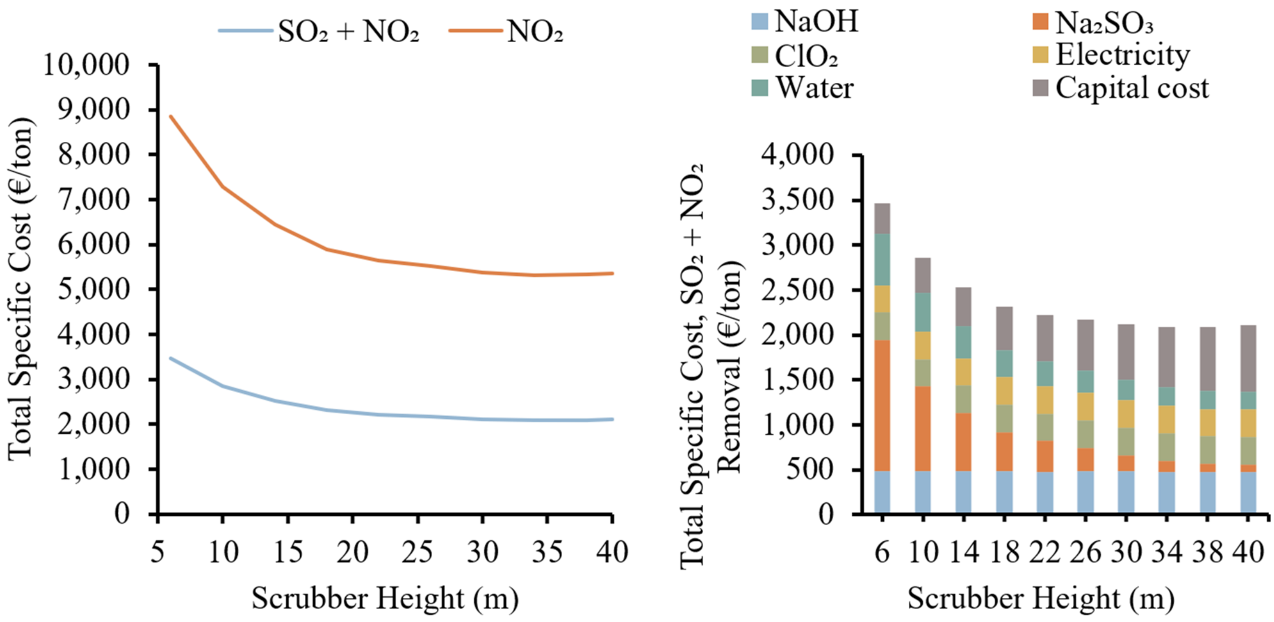

Figure 8. Impact of scrubber height on total specific cost of NO_2 and acid gas removal with constant NO_2 removal of 90% for the waste-to-heat plant case. To the left, the absolute total specific cost of NO_2 and combined SO_2/NO_2 removal in €/ton and to the right the total specific cost of combined SO_2/NO_2 removal divided by category in €/ton.

In 2015, Eelco den Boer and Maarten Hoen published an assessment report where they brought together different available investment cost figures for the installation of a SO_x scrubber on a medium-sized cruiser [34]. The capital expenditures of a scrubber for a 12 MW engine ranged from 1.2 to 2 M€ for an open loop scrubber and between 2.4 and 4.4 M€ for a closed loop scrubber. In Figure 7, we calculate the capital cost of a scrubber to be 2.2 M€. In this study, no cost is included for a waste treatment and storage solution which is part of the related increased cost for a closed loop scrubber in the compared values. The calculated cost in this study is close to the available data on installed marine scrubbers. Since the scrubber used for modelling in this study is based on an actual marine scrubber design, the numbers should be close to each other.

The operation and maintenance cost of a SO_x scrubber for marine application reported by Eelco den Boer and Maarten Hoen ranged from 0.3 to 2.5 €/MWh [34]. In this study the corresponding values can be derived from Figure 5 to be 11.3 €/MWh. When removing the NO_2 related chemicals of ClO_2 and Na_2SO_3 and excluding electricity price as this is not included in the values by Boer and Hoen, the cost drops to 7.15 €/MWh. The resulting operating and maintenance cost in this study is high compared to the values found by Eelco den Boer and Maarten Hoen. This is not unreasonable and can be explained by the cost related to co-absorption of NO_2. pH is kept higher and additional NaOH is needed to co-absorb the NO_2. It is also possible that the cost of NaOH used in this study of 400 €/ton is high compared to what shipping companies pay. In the report by Boer and Hoen, a price of 200 €/ton NaOH was assumed.

The treatment of a liquid containing both nitrogen and sulfur compounds at the volumes present in the investigated technology is not studied nor currently employed at any plant to the best of the authors' knowledge. For the time being, the maritime sector is allowed to release the wastewater while outside of ECAs why no incentives are present to introduce any cost at the moment. For the two other applications there are a couple of studies with suggestions on how to best treat the liquid where the ultimate solution would be to turn the waste into products, for instance sulfuric acid and sodium nitrate. For the case of a more conservative outlook on the eventual cost coupled to waste treatment, a more ready solution would be to inject the scrubber effluent back into the boiler. Both waste-to-heat plants and recovery boilers are subject to high particle and sulfur loads as is and should therefore be relatively insensitive to the added sulfur load. Preliminary tests performed have shown that SO_4^{2-} will remain as SO_4^{2-} when injected to the boiler in a region where the temperature is about 1000 °C and NO_2^- will be reduced to N_2. In a best-case scenario, this will only incur a minor cost from increased soot formation and depending on whether there is a flue gas condenser present or not is also an efficiency loss.

4. Conclusions

We evaluate the technical and economic performance of a co-absorption system for NO_x and SO_2 emissions control. Three cases: a small sized waste-to-heat plant, a cruise ship and a kraft recovery boiler of a medium-sized pulp and paper mill, are identified as potential targets for the technology where either process conditions or existing facilities make for an interesting business case. Based on previous experiments, a simple process set-up is established for each investigated case [7]. Each case process setup is simulated using a detailed reaction mechanism. The economic performance was investigated with investment cost estimated using the Aspen Process Economic Analyzer V11 and operational cost estimated using available data on chemical cost and simulation results. The key findings are summarized below:

1. The removal efficiencies of the co-absorption system are able to reach the level of what is currently viewed as best available technologies, with >90% and >99% removal for NO_x and SO_2, respectively.
2. The waste-to-heat plant had an estimated total specific cost of NO_2 removal of 5300 €/ton NO_2, the recovery boiler 4700 €/ton NO_2 which is relatively close to the Swedish charge on NO_2 at 4900 €/ton NO_2. The cruiser had an estimated total specific cost of NO_2 removal at 3300 €/ton NO_2.
3. The consumption of absorption chemicals (Na_2SO_3) should be significantly reduced when either an inhibitor or another absorption chemical is used, for instance $Na_2S_2O_3$.
4. The investigated technology is well suited for emission control of sources with a high share of SO_2 in the flue gas and/or sites with current availability of oxidizing chemicals and absorption chemicals.
5. The treatment of wastewater was identified as an unknown parameter in the cost evaluation.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/en14248512/s1, Table S1: Equipment list for the waste-to-heat plant, Table S2: Equipment list for the kraft recovery boiler, Table S3: Equipment list for the cruiser, Table S4: General operating conditions and design parameters for simulations and cost estimations, Table S5: Summary of Total direct cost for each investigated case together with all other resulting plant costs.

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