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## The role of energy supply in abatement cost curves for CO<sub>2</sub> capture from process industry – A case study of a Swedish refinery

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#### HIGHLIGHTS

- Techno-economic study of heat supply for partial carbon capture in process industry.
- New method to estimate heat supply cost to enhances abatement cost curves.
- Method includes multi-period optimization of heat sources; provides cost functions.
- Result: Temporal variations in residual heat impact capture cost significantly.
- Result: Site energy system of a refinery can supply heat to mitigate > 75% emissions.

#### ARTICLE INFO

# Keywords: Heat integration MILP optimization Techno-economic assessment Abatement cost curve Temporal variations Partial CO<sub>2</sub> capture

#### ABSTRACT

Carbon capture and storage (CCS) activities need to be ramped up to address the climate crisis. Abatement cost curves can help to identify low-cost starting points and formulate roadmaps for the implementation of CCS at industrial sites. In this work, we introduce the concept of energy supply cost curves to enhance the usefulness and accuracy of abatement cost curves. We use a multi-period mixed-integer linear programming (MILP) approach to find an optimal mix of heat sources considering the existing site energy system. For a Swedish refinery, we found that residual heat and existing boiler capacities can provide the heat necessary for CCS that avoids >75% of the site's  $CO_2$  emissions. Disregarding the existing site energy system and relying on new heat supply capacities instead, would lead to capture costs that are 40-57% higher per tonne of  $CO_2$ -avoided (excl.  $CO_2$  liquefaction, transport, and final storage). Furthermore, we estimated that temporal variations of heat sources (intermittent residual heat) increases the heat supply cost and emissions by 7-26% and 9-66%, respectively. The proposed method for optimization of the energy supply mix considering temporal variations of heat sources enables detailed estimates of energy supply costs for  $CO_2$  capture rates ranging from partial to full capture, and thus, improve abatement cost curves.

#### 1. Introduction

Carbon capture and storage (CCS) is an essential technology for addressing the climate crisis. Most integrative assessment models cannot identify emission pathways consistent with the 1.5 °C [1] or 2.0 °C [2] goal without large-scale CCS deployment. The current global CCS capacity is  $\sim 40$  Mt CO $_2/a$  [3], whereas the estimated required capacity by Year 2050 congruent with the 2 °C goals is  $\sim 5,600$  Mt CO $_2/a$  [4]. Thus, a fast ramp-up of CCS implementation is needed. In addition to policy

instruments, see for instance [5], the targeting of low-cost  $\mathrm{CO}_2$  sources is needed to trigger initial projects. Such sources include industrial sites with high  $\mathrm{CO}_2$  concentrations and large flows [6]. Partial capture of  $\mathrm{CO}_2$ , driven by residual heat recovery, can also help initiate early-mover projects by initially only capturing the most cost-effective share of  $\mathrm{CO}_2$ . For example, the cement plant in the Norwegian Longship project aiming for start-up in Year 2024/2025 applies partial capture, *inter alia*, because the availability of low-cost residual heat is limited [7]. Partial capture can be subsequently extended to full capture over time [8] or combined with other mitigation measures [9,10].

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Nomenclature		ICR	Iso-CRacker unit (producing low-sulphur diesel).
100	Alastania at Cart Commi	ISO	Isomerization (Iso) (chemically transforms straight
ACC	Abatement Cost Curve		hydrocarbons into branched hydrocarbons)
APC	Amine Plant Cost	KPI	Key Performance Indicator
APEA	Aspen Process Economic Analyzer	LP	Low Pressure steam
BEC	Break-Even Cost	MEA	MonoEthanolAmine; refers to an aqueous solution with 30
CAPEX	CAPital Expenditures		wt% MEA
CC	Capture Cost	MHC	Mild HydroCracker unit, desulfurizes vacuum gasoil and
CCS	Carbon Capture and Storage		converts it into lighter products and feedstock for the
CDU	Crude oil Distillation Unit		hydrocracker
CHP	Combined Heat and Power	MILP	Mixed Integer Linear Programming
CM	Cost Minimization (heat supply)	MVR	Mechanical Vapor Recompression
CRU	Catalytic Reforming Unit (for production of high-octane	NG	Natural Gas
	liquid products from naphtha distilled from crude oil)	OPEX	OPerational EXpenditures
DCC	Direct Contact Cooler that cools incoming flue gases in	NHTU	Naphtha HydroTreating Unit
	direct contact with water.	SRD	Specific Reboiler Duty; refers to heat demand for solvent
ECC	Energy supply Cost Curve		regeneration per captured CO <sub>2</sub>
EEM	External Energy Minimization (heat supply)	SRU	Sulphur Recovery Unit
EPC	Engineering, Procurement, and Construction	SSU	Sulphur Solidification Unit
FCC	Fluid Catalytic Cracking unit (cracks heavy portion of	TCR	Total Capital Requirement
	crude oil into lighter products)	TDC	Total Direct Cost
HCN	Heat Collection Network	TDCPC	Total Direct Cost with Process Contingency
HPU	Hydrogen Production Unit (via steam methane reforming)	TPC	Total Plant Cost
HSC	Heat Supply Cost	VDU	Vacuum Distillation Unit (separates heavier oils coming
HSCM	Heat Supply Cost Model	120	from atmospheric distillation).

Abatement cost curves (ACC) can help to identify the most cost-effective level of CCS implementation. At a systems level, marginal abatement cost curves can point out industrial facilities that can achieve low mitigation costs, as illustrated by Johnson et al. [11] for the process industry in Sweden or by Beiron et al. [12] for Swedish combined heat and power (CHP) plants.

Systems level analysis requires collecting a sufficient level of process data from individual sites. At the site level, ACCs have been used to determine the flue gas stacks that can achieve CCS at the lowest cost, as illustrated for refineries by van Straelen et al. [13]. This is important since there is a large heterogeneity within and between refineries, resulting in a wide spread of avoidance cost of 160-210 \$/t CO<sub>2</sub> [14]. This is caused by varying complexity of refineries and plant layouts with multiple stacks, some of which are characterized by low suitability for capture due to low flow, low concentration or high level of impurities [6,13,15]. Although CCS could only mitigate the emissions from refining operation and not the end-product emissions, CCS could reduce the refining sector's share of global CO<sub>2</sub> emissions of 4% [14] significantly. Berghout et al. [9] used ACC to evaluate combinations of mitigation options at a complex refinery including energy efficiency, CCS, and introduction of biomass feedstock. Although improved heat integration of the core refinery process was considered, Berghout et al. [9] did not include residual heat use for CCS in the cost calculations used to generate the ACC. Previous work by the authors [16] has shown that heat supply costs vary with the amount of captured CO2 and that these costs should be incorporated into the ACC to identify technoeconomically viable partial CO2 capture paths. The methodology was illustrated for partial capture at an integrated iron and steel facility. Biermann et al. [16] concluded that cost savings can be substantial when utilizing residual heat from existing site energy systems. Although not explicitly using ACCs, Johansson et al. [17] and Andersson et al. [18] found similar cost savings for refinery sites, contrary to the previous claim made by van Straelen et al. [13] that opportunities for heat integration would be marginal and would only lead to high CAPEX for typical refineries. Having attributed 45-55% of avoidance cost for refineries to utilities such as on-site CHP units, a report by IEAGHG [14] suggested that future work should focus on, inter alia, the use of residual

heat in combination with electricity import as alternative to capital cost intensive from new installations (combined heat and power plants), and the assessment of available heat generating capacity on site. It should be noted, however, that the above-mentioned works often refer to residual heat as excess heat in the conventional sense, i.e., heat that cannot be utilized for internal heat recovery. We avoid the term "excess heat" intentionally here, since residual heat that is usable for CCS would not be excess heat if CCS were seen as a necessary, thus internal, part of the process, as discussed by Eliasson et al. [19].

In this work, we developed a generic concept of energy supply cost curves (ECC) for CCS at the site level, which enables a systematic quantification of the impact of energy supply on the ACC and on the refinery operations in terms of energy efficiency and carbon emission intensity. The proposed concept improves the simplified classification of heat sources made by Biermann et al. [16] in which heat sources were ranked based on annually averaged values and existing capacities of steam generation were excluded from analysis. In line with [14], we explicitly include the use of residual heat, and the available heat generating capacity on site, as well as temporal variations of residual heat, which are likely to have an impact on cost but have not been investigated in the above-mentioned works. We propose a rigorous estimation of ECCs based on multi-period linear optimization that identifies a mix of heat sources that either minimizes costs or import of external energy, and that considers the capacity of the existing site energy system to satisfy a given heat demand. The concept of ECC is illustrated for heat supply to an amine solvent based post-combustion CO2 capture process at a Swedish refinery in a detailed case study. More specifically, the aim of this work is to:

- Derive a method to evaluate the use of residual heat and existing boiler capacity to generate steam for CCS considering intermittency of these heat sources.
- Quantify the impact of heat supply intermittency on the heat supply cost
- Incorporate ECC into site-level abatement cost curves to identify possible cost reductions at refineries with a greater degree of reliability compared to the previous studies.

 Quantify the additional direct and indirect emissions that stem from heat supply for CCS and the impact varying levels of CCS implementation has on the energy consumption (fuels and electricity) and product emission intensity of the industrial facility.

The outline of the paper is as follows: the ECC approach is presented in Section 2 together with the implementation into an optimization model. Section 3 describes the set-up of the Swedish refinery case study. Section 4 describes the results of the case study, followed by a discussion in Section 5 on variations in heat supply and the impact of heat supply cost on ACC.

#### 2. Method

Fig. 1 illustrates the framework used for the techno-economic optimisation and assessment. The energy cost curves (ECC) assess the cost of energy supply for CCS processes depending on the availability of energy in the site energy system and the amount of CO<sub>2</sub> captured. Hereinafter, the method is described in the context of heat supply for amine solvent regeneration; however, it is also applicable to other site-dependent energy flows such as process cooling. The method incorporates a heat supply cost model (HSCM), which identifies a mix of heat sources that either minimizes cost or external energy use (thus minimizes emissions) associated with heat supply. The HSCM analyses a set of pre-defined heat sources with respect to their thermodynamic properties (quantity and quality of heat) based on site data and/or process models and estimates heat supply cost, i.e., both capital (CAPEX) and operational expenditures (OPEX). These heat supply costs were included in the OPEX of the amine plant. The CAPEX of the amine plants specific to each CO2 source were based on process simulation and bottom-up cost estimation. The combination of heat supply and amine plant cost yields the capture plant cost as a function of captured or avoided CO2, as needed to compile the ACC.

## 2.1. Energy cost curve method for analysis of energy supply options at site level

Fig. 2 provides an overview of the ECC method which consists of the following eight steps:

- Site analysis for heat sources: The industrial site was analysed for indirect heat recovery opportunities via utility systems and heat potential in effluent streams in order to minimize operability constraints. Thus, an in-depth total site analysis including internal pinch analysis, see for instance [20], was intentionally avoided.
- 2. **Identification and classification of heat sources for CCS:** Typical technologies that may supply heat to the capture process are shown in Table 1 and categorized into three classes, adapted from [16]:
- Class I "residual heat"), Heat sources that may require capital expenditures (CAPEX) to be implemented but will require little operational expenditures (OPEX)/external energy costs; e.g., residual unused steam or heat in process coolers that could be used to raise steam after investment in a heat collection network,
- Class II "existing (spare) capacity"), Heat sources that require
  additional OPEX but no investment/CAPEX; e.g. existing boilers/
  equipment that could supply steam at the expense of additional
  external energy (fuel, electricity).
- Class III "new boiler capacity"), Heat sources that require both additional CAPEX and OPEX; i.e. new steam raising capacity installed at the site and fuelled by additional external energy.
- 3. Data extraction and preparation: Residual heat and existing capacity data was collected with a time-resolution that reflects typical operational variations. The capacity of existing equipment and the potential for increased production of steam was assessed considering the refinery steam demand (potential = capacity refinery demand). All process data collected was reconciled (removal of noise and

outliers) and represents the same period of time with common periods of maintenance excluded.

- 4. Thermodynamic quantification of usable heat:Based on the required reboiler temperature of the amine-solvent (here: ~120 °C for MEA) and a minimum approach temperature in the reboiler (here: 10 °C), a steam temperature of ~ 130 °C was defined. The extractable residual heat to raise steam was calculated assuming contributions to the minimum temperature difference between hot process streams and the condensate/steam according to Andersson et al. [18]. Also, steam produced at higher temperature levels (e.g., boilers) was assumed to be de-superheated for CCS purposes. Assumptions regarding efficiencies for conversion of external energy to steam (boilers, heat pumps, etc.) are presented in Section 3.1.
- 5. Cost estimation via bottom-up and/or top-down approach: Both capital and operational expenditures were considered in estimating the cost of heat supply for each heat source. Technologies that require installation of equipment were estimated via a mixed bottom-up and top-down approach (see Section 2.4) depending on the heat source and the availability of cost data (see Section 3.1).
- 6. **Quantification of external energy demand and emissions:** Scope 1 and Scope 2 <sup>1</sup> emissions were quantified from the heat sources due to consumption of external energy (fuel, electricity).
- 7. Optimization of technologies: A mix of heat sources was determined to meet the given constant, annual heat load of the capture unit using multi-period optimization (hourly resolution). For periods when residual heat sources (Class I) are fully used, the optimization function identifies the best option (Class II or Class III) that can operate and compensate for intermittencies in residual heat.
- 8. Comparison of KPI's and assembly of cost curves: Key performance indicators (KPI) were calculated for the identified mix of heat sources according to Eq.'s 3–5. Step 7 was performed for a range of constant steam demands, covering partial to full CO<sub>2</sub> capture, to enable the generation of the ECC.

The heat supply cost C, the amine plant cost APC, and the capture cost  $CC_{\rm avoid}$  (both per unit of captured and avoided  $\rm CO_2$  are used, the latter is illustrated here) – cf. Fig. 1 – were calculated according to Eq.'s (1), (2), and (3), respectively. Note that pre-processing and  $\rm CO_2$  collection,  $\rm CO_2$  liquefaction and handling (on-site storage/ $\rm CO_2$  terminal), and shipping and storage costs were not included in this work. Two additional performance indicators were defined to assess the impact that CCS has on the energy efficiency of the production  $\eta_{\rm production}$  and the product emission intensity of the refinery  $e_{\rm product}$ , see Eq. (4) and (5), respectively. To compare the output of the HSCM generated by the two objective functions (minimize cost or energy), the break-even cost ( $BEC_{\rm CO_2}$ ) between the two solutions is calculated according to Eq. (6).

$$HSC = \frac{\sum_{\text{heat sources i}} (CAPEX + OPEX)_{i}}{\sum_{\text{heat sources i}} (m_{\text{steam,CCS}})_{i}} \left[ \frac{\epsilon/a}{t \text{ steam/a}} \right]$$
(1)

$$APC = \frac{\sum_{\text{AP j}} (CAPEX + OPEX)_{j}}{\sum_{\text{AP j}} (m_{\text{CO}_{2}, \text{captured}})_{j}} \left[ \frac{\epsilon/a}{t \text{ CO}_{2}/a} \right]$$
 (2)

$$CC_{\text{avoid}} = \frac{(CAPEX + OPEX)_{\text{AP}} + (CAPEX + OPEX)_{\text{HS}}}{m_{\text{CO}_2, \text{captured}} - m_{\text{CO}_2, 2, \text{AP}} - m_{\text{CO}_2, 1+2, \text{HS}}} \left[ \frac{\epsilon/a}{\text{t CO}_{2, \text{avoided}}/a} \right]$$
(3)

$$\eta_{\text{production}} = \frac{\sum_{\text{p}} E_{\text{product,p}}}{E_{\text{refinery,input}} + E_{\text{AP}} + E_{\text{HS}}} \times 100 \ [\%]$$
 (4)

<sup>&</sup>lt;sup>1</sup> Scope 1 are direct greenhouse gas emissions from sources owned by a company. Scope 2 accounts for indirect greenhouse gas emissions from the generation of purchased energy, e.g., electricity. See [71].

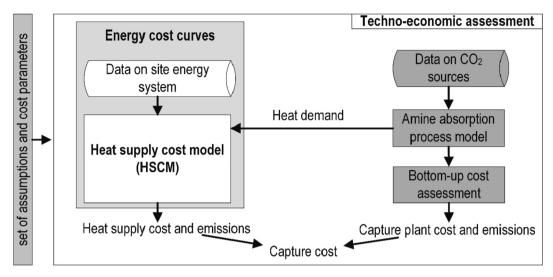


Fig. 1. Overview of the applied methodological framework. The proposed method of energy cost curves is the focus of the work and incorporates the heat supply cost model (HSCM).

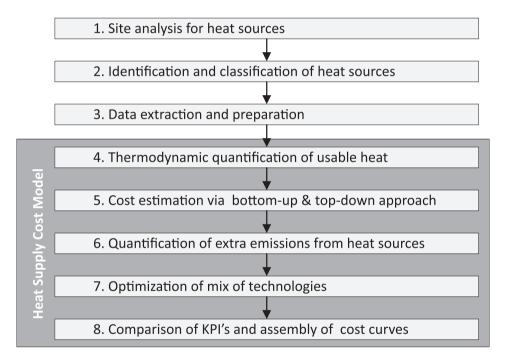


Fig. 2. Overview of the method to generate energy cost curve for the analysis of energy supply options for CCS at site level and the automation of step 4–8 with the heat supply cost model.

$$e_{\text{products}} = \frac{emissions_{1+2,\text{Refinery}} - emissions_{\text{avoided,AP+HS}}}{\sum_{ip} E_{\text{product,p}}} \begin{bmatrix} \text{kg CO}_2/\text{a} \\ \text{GJ/a} \end{bmatrix}$$
 (5)

$$BEC_{\text{CO}_2} = \frac{HSC_{\text{CM}} - HSC_{\text{EEM}}}{emissions_{1+2, \text{ CM}} - emissions_{1+2, \text{EEM}}} \begin{bmatrix} \frac{\epsilon}{4} \\ \frac{1}{4} \end{bmatrix}$$
(6)

The indices in Eq.'s (1)-(6) are,

1	
HS	Heat supply
i	Heat source i
j	Amine plant j (1 per source)
p	Refinery product p
E	Energy
AP	Amine capture plant(s)
1	Scope 1 emissions
2	Scope 2 emissions
CM	Cost minimization
EEM	External energy minimization.

The energy input into the refinery  $E_{\rm refinery,input}$  includes the crude oil, LNG, and electric power. The energy demand by the amine plant  $E_{\rm AP}$  includes the power demand for pumps, fans and air cooling. The energy demand for heat supply  $E_{\rm HS}$  includes power and natural gas demand.

#### 2.2. Heat supply cost model

The HSCM automizes the calculations of Steps 4–8 (Steps 1–3 are independent of input values such as steam temperature or economic parameters) in the above proposed method (cf. Fig. 2). The model workflow is illustrated in Figure S. 1 (Supplementary Material S.2). Note that the  $\rm CO_2$  capture unit is not part of the optimization to simplify the problem given the scope of the paper (cf. Fig. 1). The model derives *linear* cost and energy functions depending on techno-economic

Table 1

Classification of CCS heat supply sources at industrial facilities."Intermittent" denotes strong, irregular fluctuations coupled to the refinery process; "Steady" denotes small fluctuations, and in principle following the operation/production load of the industrial process; "Variable" means manageable in the sense that steam generation could be adjusted fairly independently of the operation/production load of the industrial process.

Class of heat supply	Examples of heat supplying technologies	Character
I Residual heat	Vented steam Excess gas flaring (other than for safety purposes, start-up, shut down)	Intermittent Intermittent
	Heat recovery steam generators (using e.g. hot flue gases) Heat collection network (steam raising, hot water collection) Heat collection with heat pumping Heat recovery from CO <sub>2</sub> conditioning unit	Intermittent/ variable ~Steady ~Steady ~Steady
II Existing capacity	Switching comp./pump drive from steam turbine to electric motor;	Variable
(external energy)	Turbine back-pressure operation (condensing turbine is installed) Increase in load of steam boilers (often fossil fuel such as oil or gas)	Variable Variable
III New heat capacity (external energy)	Installation of new steam boilers (fossil, biomass, electric) Installation of CHP plants (fossil, biomass)	Variable Variable

parameters as input to the multi-period optimization function (Step 7) that finds an optimal technology mix (among the available set of technologies) with the objective to either minimize cost of heat supply or minimize external energy (purchased energy carriers for heat supply). The optimization considers a set value (constant steam demand) and constraints (maximum capacity of each technology). For the description of the optimization problem, the following index sets notations are used:

 $P = \{p\}$ : index set of discrete time periods,

 $T = \{t\}$ : index set of available technologies.

The objective of the optimization is to minimize the total annualized cost (TAC) of heat supply which comprises annualized investment cost ( $CAPEX_a$ ) and operating cost (OPEX), which depend on the chosen technologies and are formulated in Eqs. (7)-(9). The decision variables, x, are explained and explicitly named in the following paragraphs.

$$minTAC(x) = CAPEX_a(x) + OPEX(x)$$
(7)

$$CAPEX_{a}(x) = \sum_{t=1}^{T} CAPEX_{a,t}(x)$$
(8)

$$OPEX(x) = \sum_{t=1}^{T} OPEX_{t}(x)$$
(9)

The CAPEX for each technology t in T ( $CAPEX_{a,t}$ ) are described by a general linear cost model, see Eq.(10), in which  $q_{t,cap}$  describes the installed thermal capacity of technology t (e.g. in MW) and the parameters  $m_{CAPEX,t}$  and  $b_{CAPEX,t}$  are positive numbers and are determined for each technology t prior to the optimization. Consequently, the installed thermal capacity of each technology t in T ( $q_{t,cap}$ ) can be interpreted as a decision variable which can be manipulated by the numerical solver to identify the optimal solution. To further account for the possibility that a technology t in T is not installed, i.e.,  $q_{t,cap} = 0$ , binary variables  $y_{CAPEX,t}$  are introduced (to avoid fixed costs of  $b_{CAPEX,t}$  when a technology is not installed). Additional constraints are necessary to invoke that the binary variables are exactly 0 when the decision variables are 0 (see Eq. (11)). These so-called Big-M constraints are characterized by the Big-M parameters which have to be large values exceeding the maximum (expected) value of the corresponding decision variable ( $q_{t,cap}$ ).

$$CAPEX_{a,t}(q_{t,cap}, y_{CAPEX,t}) = m_{CAPEX,t} \cdot q_{t,cap} + b_{CAPEX,t} \cdot y_{CAPEX,t}$$
(10)

$$q_{t,cap} - BigM^*(1 - y_{CAPEX,t}) \le 0 \tag{11}$$

The OPEX for each technology t in T ( $OPEX_t$ ) is the sum of the operating cost of technology t for all discrete time periods p in P, see Eq. (12), where the OPEX in each discrete time period p ( $OPEX_{t,p}$ ) is described as by a general linear model, see Eq.(13). Therein,  $q_{t,p}$  describes the thermal contribution of technology t for time period p to satisfy the heat demand during time period p ( $Q_{\text{dem,p}}$ ) and is a decision variable. The parameters  $m_{\text{OPEX,t}}$  and  $b_{\text{OPEX,t}}$  are positive numbers which are determined prior to the optimization. Similarly to the calculation of CAPEX, a technology t in T may not operate during time period p, i.e.  $q_{t,p} = 0$ . Again, binary variables  $y_{\text{OPEX,t,p}}$  are introduced and additional BigM constraints (see Eq. (14)) are formulated. For clarification, the numerical solver will always strive to set the binary variables  $y_{\text{CAPEX,t}}$  to 0 as this decreases the values for  $CAPEX_{a,t}$  (compare Eq. (10)). However, by means of Eq.11, this is only possible for the case that the corresponding decision variable ( $q_{t,cap}$ ) is also 0.

$$OPEX_{t}(x) = \sum_{p=1}^{P} OPEX_{t,p}(x), \forall t \in T$$
(12)

$$OPEX_{t,p}(q_{t,p}, y_{OPEX,t,p}) = m_{OPEX,t} \cdot q_{t,p} + b_{OPEX,t} \cdot y_{OPEX,t,p}$$
(13)

$$q_{t,p} - BigM \cdot (1 - y_{OPEX,t,p}) \le 0 \tag{14}$$

The installed thermal capacity of each technology t ( $q_{t,cap}$ ) should ideally be only as large as the maximum experienced thermal contribution of each technology t for any time period p in P ( $q_{t,p}$ ), which is formulated as mathematical constraint in Eq. (15). Another constraint is the heat demand during time period p ( $Q_{\text{dem},p}$ ) which must be satisfied for each time period according to Eq. (16), which is set as a constant value in this work. The decision variables  $q_{t,cap}$  and  $q_{t,p}$  are bound to be positive real numbers according to Eq.(17).

$$q_{t,\text{cap}} \ge q_{t,p} \forall p \in P, \forall t \in T, \tag{15}$$

$$Q_{\text{dem},p} = \sum_{t=1}^{T} q_{t,p} \forall p \in P$$
 (16)

$$q_{t,\text{cap}}, q_{t,p} \ge 0 \forall p \in P, \forall t \in T.$$
 (17)

In summary, the defined optimization model can be classified as a multi-period mixed integer linear programming model (MILP). To minimize the annual external energy demand, an analogous model is defined. However, since the energy demand is only dependent on the demand in each discrete time period, no contribution analogous to the CAPEX contribution (Eq. (7)) is required. Additionally, the definition of integer variables is not necessary since the linear model used to describe the external energy demand in each discrete time period does not include a constant term, i.e., no term analogous to  $b_{\text{CAPEX},t}$  or  $b_{\text{OPEX},t}$  is included.

#### 2.3. Process modelling of CO2 absorption

The modelling of  $\mathrm{CO}_2$  capture was carried out using Aspen PLUS V11 software. The standard capture plant configuration, as shown in Fig. 3, was considered. The solvent is an aqueous monoethanolamine (MEA) solution with a concentration of 30 wt%. The model was based on previous work [8,21] and was revised with respect to property method and reaction sets. The liquid properties were estimated using the ENRTL-RK method, whereas the vapor properties were estimated using the PC-SAFT equation of state. All columns were modelled by rate-based calculations, assuming counter-current flow of vapor and liquid. Mass transfer coefficients and interfacial areas in the packings were predicted

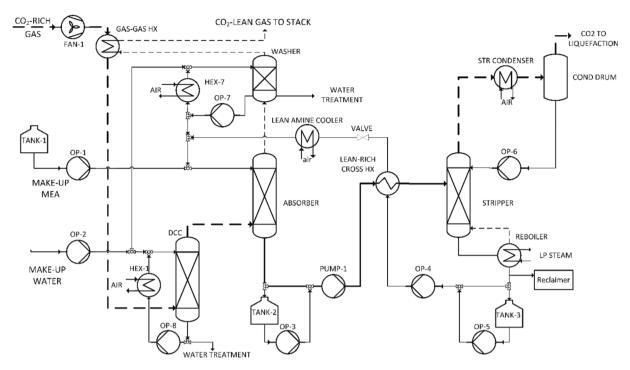


Fig. 3. Process flow diagram of the standard configuration amine capture process with all major components included in the cost estimation for the capture plant.

using the correlations by Bravo et al. [22]. The holdup correlations by Stichlmair et al. [23] were used. Heat transfer coefficients were obtained via the Chilton and Colburn analogy [24]. Two reaction sets were implemented – one for the absorber and one for the stripper that differ in the reaction constant for the reverse carbamate reaction [25]. The model performance was verified against experimental data presented by Faramarzi et al. [26], and specific reboiler duty calculated by the model deviated from the experimental data by  $\sim$  3%. For the direct contact cooler (DCC), absorber and the stripper, the design-mode was active, meaning that the column diameter was calculated assuming a flood approach of 80% (for a specified value of the packing height). The specific reboiler duty was minimized for each capture plant by varying the lean loading (reboiler temperature) and the solvent flowrate to achieve 90% capture. Note that 90% capture is commonly assumed to be the cost-optimum capture rate in many academic studies, although recent studies indicate that higher capture rates may in fact be viable, see [8,27-29]. Details on the design parameters and assumptions on consumables (water, chemicals) can be found in Supplementary Materials S.5.1.

#### 2.4. Cost estimation method

#### 2.4.1. Capital cost

Based on latest guidelines for cost evaluation of CCS [30,31], the cost estimation followed the economic assumptions shown in Table 2. Fig. 4 illustrates the conducted combined top-down and bottom-up cost estimation method adapted from [32,33]. Bottom-up implies the estimation of direct cost for individual pieces of equipment based on equipment design obtained from process modelling with cost information either directly derived from Aspen Process Economic Analyzer or indirectly (literature). In contrast, top-down implies the use of literature values for an entire unit, which is often represented as engineering, procurement, and construction cost (EPC). Both approaches use (the same) respective cost escalation factors (cf. Table 2 and Fig. 4).

The automated, bottom-up cost estimations within the HSCM were based on detailed cost functions which were regressed from multiple cost evaluations for specific equipment using the Aspen Process Economic Analyzer (APEA), similar to [34,35]. These functions for direct

cost (equipment and installation) without process contingency for required equipment are listed in Supplementary Materials S.4.1. The top-down assessed cost functions of mechanical vapor recompression [36–38] and the new electric or gas-fired boilers [38] are listed in Supplementary Materials S.4.2.

The capital costs of the amine plants were estimated from approximate, bottom-up, direct-cost functions which were obtained via regression from literature [16,32,39–43,70] and from APEA. These and can be found in Supplementary Material S.5.2. Compared to detailed cost functions for heat supply equipment, the amine plant cost curves have lower accuracy and only follow one sizing parameter. However, they represent the order-of-magnitude of capital cost and the expected scaling behavoir, which was deemed sufficient for the scope of the work, i.e., the study of heat supply cost in the context of other capture cost. An exemplary equipment list for the capture unit is shown in Supplementary Material S.5.3. To generate the capture cost curve, the total CAPEX of each plant is then scaled against captured CO<sub>2</sub> using power law functions derived by Garðarsdóttir et al. [44] assuming that the design, and thus specific energy requirements, are maintained.

#### 2.4.2. Operational cost

For the heat sources, operational cost included fixed (maintenance, insurance) and variable cost (fuel/electricity, auxiliary electricity, other variable O&M cost assumed for boiler operation). Detailed assumptions used in the Case study are described in Table 5 in Section 3.1. For the capture units, the fixed OPEX also included labour costs to operate the plant (6 operators, 1 engineer). The variable OPEX included the auxiliary electricity for pump operations and air-cooling of the amine process as well as other consumables (process make-up water, chemical cost comprising make-up solvent, caustic soda and reclaimer sludge disposal), see Table 2.

**Table 2**Default economic parameters for the heat supply cost model.

CAPEX			
Cost year	_	2018	
Lifetime	Years	25	
Cost index	_	PCD	Price index reflecting central
			European chemical plants [46]
Location factor	_	0.995	Sweden relative to Netherlands
			[39] (adjusted to cost 2018)
Construction	Years	3	Interest during construction, see
			[33]
Discount rate	%	8	Average discounted cash flow rate (no inflation)
Annualization factor	TCR/	10.67	Calculated according to [39]
	CAPEX		
Process contingencies	%TDC	5–15	5% heat supply equipment; 15%
			capture plant equipment & class B
			exchangers; assessed according to
			[31,47,69]
Indirect cost	%	25	[14]
	TDCPC		
Project contingency	%EPC	40	assessed according to [47]
Owner's cost, spare	%TPC	9.5	[14]
parts and			
modifications			
OPEX			
Availability (CCS plant & heat supply)	h/a	8500	site specific
Maintenance + insurance	%TPC/a	4.5	annual cost % total plant cost [33]
Labour cost	k€ <sub>2018</sub> /a	411	6 operators, 1 engineer; Swedish
	2018/		labour cost based on average wages
			[48], escalated to full-burdened
			cost [49].
Natural gas price	$\epsilon_{2018}$	41.4	Swedish industrial consumers
	MWh		2018, including taxes and network
			charges, no VAT [50]
Electricity price	€2018/	58.40	Swedish non-household consumers
7.1	MWh		2018, including taxes and levies, no
			VAT [50]
MEA price	€ <sub>2018</sub> /t	1700	[16]
Reclaimer sludge	€/t	300	[16]
disposal cost			
Caustic soda (NaOH)	€/t	400	[32]
price			
Process water price	€/m³	6.65	[32]

Table 3  ${\rm CO_2}$  sources at the Lysekil refinery; for unit abbreviations see Nomenclature section; Stacks 4 and 6 excluded due to low flue gas flows.

			U		
		Stack 1	Stack 2	Stack 3	Stack 5
Sources of flue gas	Unit	SRU, CDU, VDU, fired boilers, incin.	CRU, MHC, SSU, ISO, NHTU	FCC	HPU
Flue gas flow (dry)	kNm³/ h	379	268	87	130
Temperature	°C	144	160	216	160
$CO_2$	vol.%	7.0	7.0	13.0	20.4
$N_2$	vol.%	73.0	73.4	78.1	61.1
H <sub>2</sub> O	vol.%	13.0	13.0	7.0	17.1
$O_2$	vol.%	7.0	6.6	1.9	1.4
Operating hours per annum	h/a	8,585	8,585	8,500	8,500
Contribution to emissions baseline	kt CO <sub>2</sub> /a	508	359	202	535

**Table 4**Design and emission intensities for the case study including HPU scenario analysis and analysis of abatement cost curves ACC.

Parameter	Unit	HPU scenario	ACC	Comment
Steam temp.	°C	131	131	Saturated; dTmin = 10 °C in reboiler
Reboiler temp.	°C	121	121	From CO <sub>2</sub> capture modelling
Capture rate	%	90%	90%	90% of 2018 annual emissions from resp. stacks
CO <sub>2</sub> -captured	t CO <sub>2</sub> /h kt CO <sub>2</sub> /a	56.65 482	1–170 8–1442	max.value refers to 90% CO <sub>2</sub> from all four major stacks.
Specific reboiler duty	MJ/kg CO <sub>2</sub>	3.34	3.34 – 3.93	From CO <sub>2</sub> capture modelling; constant load for 8500 h/a
Electricity emission intensity	gCO <sub>2eq</sub> / kWh	47	47	Swedish grid CO <sub>2</sub> intensity 2013 [54]
Natural gas greenhouse gas intensity	gCO <sub>2eq</sub> / MJ	65.9	65.9	EU28 natural gas incl. distribution emissions [55]

#### 3. Case study - Swedish petroleum refinery

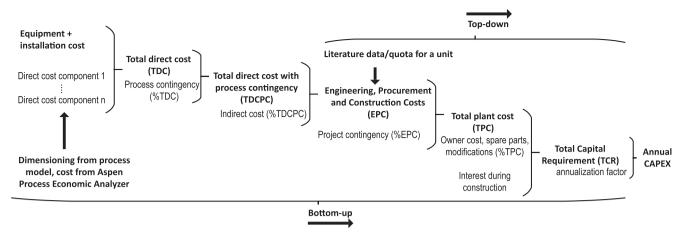
The case study was based on the "complex" refinery in Lysekil, Sweden, with a capacity of 11.4 Mt crude oil/a (220,000 bpd) with estimated direct and indirect emissions of about 1.70 Mt CO2/a and 0.037 Mt CO<sub>2</sub>/a, respectively. The product volume and production efficiency for the reference year 2019 were 12,285,000 m<sup>3</sup> and 89.37%, respectively [51,52]. Fig. 5 shows the studied system, the included heat sources, and the applied method for each heat source in yellow. The quantification of residual heat and existing capacity was based on Year 2018 data for the refinery steam system using a stream data set for 36 selected product coolers (air coolers). The potential of heat recovery from hot flue gases was assessed as too limited and thus, excluded. Also, as mentioned in Section 2.1, additional internal heat integration between process units assuming a total site analysis approach, as well as additional heat recovery from other effluent streams, were not included in the analysis. The scope of heat recovery in the case study was limited to low-pressure steam raising. Thus, heat recovery from the CO2 capture plant at lower temperatures, e.g., via hot water collection, was not considered. The major flue gas stacks of the refinery are shown in Table 3 – most CO<sub>2</sub> emissions are generated in the hydrogen production unit (HPU) with a CO2 concentration of 20 vol.%wet. Capture of 90% of yearly CO<sub>2</sub> emissions from all four major stacks would amount to 1442 kt CO<sub>2</sub>/a, here denoted as full capture.

The Case study is divided into four parts:

Scenario analysis – capture from HPU:  ${\rm CO_2}$  capture from the HPU flue gas (steam methane reforming) is studied to illustrate the variability of residual heat sources and their impact on cost and emissions of heat supply.

Analysis of heat supply cost curves (ECC): The impact of capture rate on heat supply in terms of cost and emissions is illustrated by running the HSCM in a range of 1-173 MW of constant heat supply. Analysis of  ${\bf CO_2}$  abatement at site: The impact of heat supply on ACC,  ${\bf CO_2}$  abatement at site, and the defined KPIs on production efficiency and product emissions intensity is illustrated.

 $<sup>^2</sup>$  The refinery has a Nelson Complexity Index of 10 [72], and has visbreaker, fluid catalytic cracker, mild hydrocracker, and hydrocracker upgrading units that convert a large fraction of residual fuel oils to lighter, more valuable products.



**Fig. 4.** Capital cost estimation method. Arrows indicate entry point and direction of cost escalation. Illustration adapted from [32,34,45]

Table 5
CAPEX, OPEX and energy contributions for the linearized cost and energy functions for each heat source. General efficiencies considered include adiabatic pump/compressor and mechanical efficiencies of 80% and 95%, respectively.

Heat source (Class)	CAPEX	OPEX & Energy			Max. capacity determined by
Vented steam (I)	No <sup>(1)</sup>	No			Steam data
Heat recovery steam generators (I)	No <sup>(1)</sup>	Power to pump boiler feed: Variable O&M:	0.0014 0.001	kW/kW <sub>heat</sub> €/kWh <sup>(2)</sup> <sub>heat</sub>	Steam data; operational factor
Heat collection network (I)	Bottom-up: Exchangers Cond. pump Piping Cond. tank	Power to pump condensate:	4.8e- 04	kW/kW <sup>(3)</sup> <sub>heat</sub>	Process cooler data; operational factor
	Top-down: MVR compressor	Power to compress steam:	0.1117	kW/kW <sub>heat</sub>	
Switchable drives (II)	No (1)	Power to drive electric motors instead of steam turbines:	0.5936	kW/kW <sub>heat</sub>	Steam data
Steam boilers (II)	No <sup>(1)</sup>	Natural gas use:	1.176	kW/kW <sup>(4)</sup> <sub>heat</sub> kW/ kW <sup>(2)</sup> <sub>heat</sub> €/kWh <sup>(2)</sup> <sub>heat</sub>	Steam data
		Power use:	0.0014	kW/kW <sub>heat</sub>	
		Variable O&M:	0.001	€/kWh <sub>heat</sub>	
New steam boiler (III)	Top-down	Natural gas use:	1.176	kW/kW <sub>heat</sub>	Practically unlimited;
	-	Power use:	0.0014	kW/kW <sub>heat</sub>	modularized > 50 MW
		Variable O&M:	0.001	€/kWh <sub>heat</sub>	
New electric boiler (III)	Top-down	Power use ('fuel', (5))	1.01	kW/kW <sub>heat</sub>	Practically unlimited;
	-	Power use (auxiliary):	0.005	kW/kW <sub>heat</sub>	modularized > 50 MW
		Variable O&M:	0.0005	€/kWh <sup>(2)</sup>	

<sup>(1)</sup> It was assumed that the existing steam system has sufficient capacity to deal with steam flows from residual heat and existing capacity and can transport steam to the reboiler via the existing network.

**Sensitivity analysis:** The heat supply cost and  $CO_2$  avoidance cost for capture from the HPU is compared to all-stack capture, together with a sensitivity analysis of key techno-economical parameters.

#### 3.1. Assumptions for the case study

The assumptions for the case study are shown in Table 4. A site specific assumption is the use of air-coolers for process cooling, which leads to an electricity demand of  $0.07-0.18~\mathrm{MJ/kg~CO_2}$  based on the design approach by Towler and Sinnott [53] and temperatures in the MEA process not lower than 38 °C. For simplicity, it was assumed that each flue gas stack has a separate capture unit, i.e., no clustering of gases or stripper columns was assumed, and that the merit order of the stacks (cf. Table 3) is: Stack 5 (HPU), Stack 3 (FCC), Stack 2, Stack 1. The merit order is based on CO<sub>2</sub> concentrations, followed by impurities (SO<sub>x</sub>), and

then flow. Note that emissions from natural gas combustion for heat supply were not considered for capture.

#### 3.2. Identified heat sources

The heat sources identified are described briefly below. Fig. 6 shows the variability of Class I and Class II heat sources. Table 5 lists the considered cost and energy contributions for each heat source. Steam from Class I + II heat sources above the CCS temperature level (131  $^{\circ}$ C) is transported in existing steam networks and throttled and desuperheated prior to the reboiler, thus some Class I and II heat sources have no CAPEX contributions. The heat collection network is a newly installed, separate network that transports low-pressure (LP) steam below the operating pressure of existing steam networks. The steam from all heat sources was assumed to be collected in a steam drum (residence time 10 s) and sent to the reboiler via piping (200 m). The

<sup>(2)</sup> literature values, taken from [38].

<sup>(3)</sup> calculated, varies with e.g. steam temperature, length of piping, etc.

<sup>(4)</sup> a boiler efficiency of 85% is assumed, based on data analysis by [59].

<sup>(5)</sup> electric boiler efficiency of 99% was assumed based on [60].

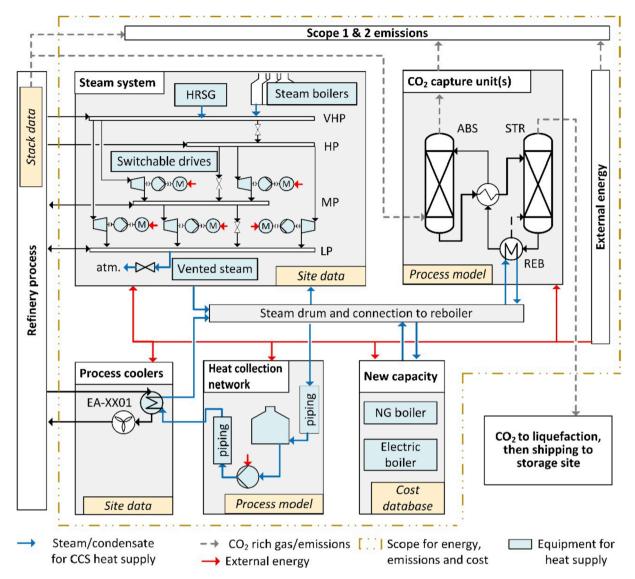


Fig. 5. Overview of the studied systems consisting of CO<sub>2</sub> capture units and heat supply. Units connected to blue arrows and highlighted in light blue are new capacity, i.e. units that have to be installed or have to increase their capacity. The yellow fields denote the method applied in the techno economic evaluation. For abbreviations see the Nomenclature section.

resulting cost for both items is hereafter referred to as "connection cost".

**Vented steam (Class I):** LP steam ( $\sim$ 150 °C, 4.8 bara) that is vented or condensed due to (1) excess of LP steam from steam turbines (frequent, short periods), or (2) excess of refinery fuel gas being mitigated by producing steam in boilers (on hot days).

Heat recovery steam generators (Class I): Two heat recovery steam generators with a maximum production capacity of high-pressure steam ( $\sim$ 390 °C, 40 bar) of 24 and 29 t/h, respectively have spare capacity due to (1) an excess of steam in the system, (2) reduced intake of fuel gas (leading to lower flue gas flows), and (3) constraints on stack temperature (acid dew point) leading to a partial bypass of the heat recovery steam generators. The difference in load between actual production and maximum production is considered as potential heat source for CCS. This load difference is multiplied with an operational factor (see Figure S.2 in Supplementary Material S.1) that represents the utilization of the refinery (based on fuel consumption) to better reflect the refinery operation and the actual possible increase in load.

Heat collection network (HCN, Class I): The HCN raises LP steam (131 °C sat.) from process streams that are currently cooled by air-coolers. The LP steam is piped to the designated reboiler location in a HCN similar to the work of Ali et al. [56], see Fig. 5. The existing air

coolers cover the remaining cooling load of the process stream.

Mechanical vapor recompression (MVR) was considered as heat pump option for the HCN, see [18]. It recompresses LP steam (102  $^{\circ}$ C sat.) to temperature levels suitable for CCS (131  $^{\circ}$ C sat.). Note that the HCN with MVR is an alternative to the regular HCN, i.e. all steam was either raised above or below the reboiler temperature in a centrally placed MVR. Details on the design of the HCN and description of simple process models can be found in Supplementary Material S.3.

**Switchable drives (Class II):** Some of the compressors and pumps on site can switch their drives from steam turbine to electric motors. Based on annual data, the amount of retrievable heat from switching drive mode was assessed based on data provided in [57,58]. The steam bypassing the turbine was assumed to be de-superheated, thus increasing the mass flow of LP steam.

Existing natural gas boilers (Class II): Three natural gas (NG) fired steam boilers are currently operated at site with a maximum production capacity of high pressure steam ( $\sim$ 390 °C, 40 bar) of 90 t/h each [59]. Based on steam system data, the possible increase in load was estimated. Two of the three boilers were assumed to be available; one was kept on minimum load as redundancy.

New boiler capacity (Class III): New LP steam generating capacity

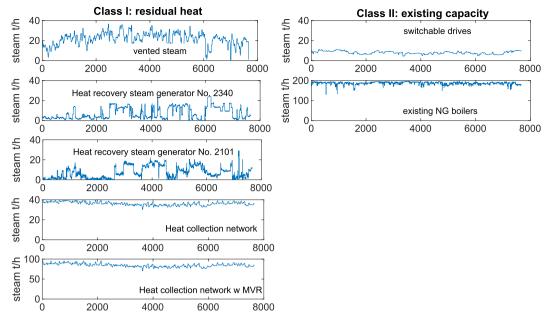


Fig. 6. Example of data-derived residual heat and existing capacity for a steam temperature of 131 °C. "w MVR" denotes with mechanical vapor recompression.

may be installed in the form of NG fired boilers or electric boilers. Combined heat and power production was excluded from the scope of this work $^3$ . Details on the boilers can be found in Table 5 in Section 3.1 below.

#### 3.3. Sensitivity analysis

A one-at-a-time, local sensitivity analysis [61] was conducted including the sensitivity to: 1) selected technical parameters in form of steam temperature and specific reboiler demand, 2) economic parameters in form of discount rate (CAPEX) and energy prices, and 3), pa-

**Table 6**Parameters and their range included in the one-way sensitivity analysis.

	Ü		
Parameter	Unit	Range	Reference/Comment
Steam temperature	°C	125 – 150	Low: $\Delta T_{min} = 4$ °C; High: steam from HCN can be transported in existing network (~150 °C)
Specific reboiler duty	%	-10 - +10	Percentual change as compared to modelled, CO <sub>2</sub> -source specific value. Covers model inaccuracies/simpler process modifications
CAPEX - Discount rate	%	4 – 15	Upper bound of 15% gives similar CAPEX as a lifetime of 10 years at 8% discount rate
Electricity price	€/MWh	29.1 – 87.3	+/- 50% of base case value
Gas price	€/MWh	20.7 – 62.1	+/- 50% of base case value
Emissions intensity power grid	gCO <sub>2eq</sub> / kWh	0 – 300	From dedicated renewable to $\sim$ EU average [62]
Emissions intensity gas	$\mathrm{gCO}_{\mathrm{2eq}}/$ MJ	4.7 – 65.9	4.7: corresponds to 95% savings of EU fossil fuels (94 g/MJ); [55]

rameters on the emission intensity of external energy (energy system). These parameters and their range of values are shown in Table 6.

#### 4. Results

#### 4.1. Impact of intermittent residual heat and existing capacity

Fig. 7 shows the heat load distribution over the course of a year for

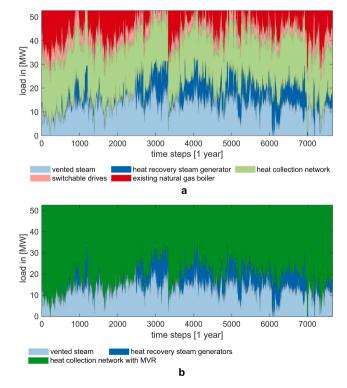
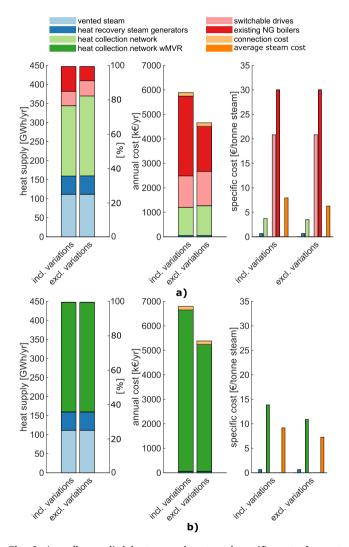


Fig. 7. Load distribution of a cost-minimized (a) and external-energy-minimized (b) heat supply of 52.56~MW of steam at  $131~^{\circ}C$  for single-stack capture from the HPU. The order of merit of the heat sources (bottom to top) is determined by either lowest cost or emissions respectively. Notations: "NG", natural gas; "wMVR", with mechanical vapor recompression;

<sup>&</sup>lt;sup>3</sup> Note that power production, e.g., with CHP plants, was excluded intentionally, since the profit from power generation may lead to improper suggestions of building large-scale power plants at a refinery site given sufficiently high electricity prices. See Berghout et al. for a discussion on the impact of the sale of excess electricity [9].

both cost-minimum (CM) and external-energy-minimum (EEM) heat generation. The target steam demand for single-stack capture from the HPU flue gas can be met with residual heat and existing capacity sources (Class I and II) exclusively, i.e., no new steam generating capacity (Class III) is required. Fig. 7 illustrates the impact of heat source intermittency and the complementing heat sources needed. For the CM solution (Fig. 7 a), the existing steam boilers (Class II) have a sufficient capacity to complement the intermittent residual heat. For the EEM solution (Fig. 7b), a heat collection network with MVR (Class I) is instead installed to manage the variations, since the switchable drives and existing steam boilers require substantial amounts of external energy, which is not selected by the EEM optimization.

Fig. 8 shows the share of annually supplied heat, the associated annualized cost (CAPEX, fixed OPEX, and variable OPEX) and the specific cost per heat source for both the CM and EEM solutions. For the CM solution (Fig. 8a), the largest heat source is the HCN (without MVR), which accounts for  $\sim$  41% of the supply at cost < 4  $\ell$ /t steam. The natural-gas-fired steam boilers represent  $\sim$  10% of the supply yet show the highest specific cost. Intermittent residual heat (vented steam and



**Fig. 8.** Annually supplied heat, annual cost, and specific cost of a cost-minimized (a) and external-energy-minimized (b) heat supply of 52.56 MW of steam at 131  $^{\circ}$ C for 90% single-stack capture from the HPU flue gas. The optimization was either run including typical variations (process data) or on annually averaged values. Notations in figure legend: "wMVR", with mechanical vapor recompression; "connection cost", cost for piping (reboiler to existing steam network) and steam drum; "average steam cost", specific cost of the entire mix of heat sources incl. connection cost.

heat recovery steam generators) shows little or no cost. For the EEM solution (Fig. 8b), the HCN with MVR dominates the amount of supplied heat ( $\sim$ 64%) and the cost.

Fig. 8 also shows the impact of intermittency of the residual heat on steam cost. Running the CM solution for an annual average of the variations yields 26.4% lower steam cost than including process data derived variations. This is because avoided intermittency leads to a better utilization of the installed HCN, which reduces the need of natural gas for peak-demand, saving 66% in emissions. Similarly, the EEM solution run on annual averaged values leads to cost savings of 26.2%. The installation of excess capacity of the HCN with MVR to cope with intermittency is avoided. However, emissions savings are small (0.01% of the run with typical variations) since approximately the same average heat load is provided.

Table 7 summarizes and compares the CM and EEM solutions. The EEM solution is  $\sim 15\%$  more expensive than the CM solution due to a large HCN with MVR – which uses 12 instead of 5 heat exchangers at a total capacity of 51 MW instead of 23 MW. In terms of emissions, the CM solution emits 14 times as much CO $_2$  as the EEM solution. Still, this is only  $\sim 4\%$  of the amount captured (482 kt CO $_2$ /a). Note the break-even CO $_2$  price of  $\sim 50~\text{€/t}$  CO $_2$  at which the EEM solution reaches the same cost-level as the CM solution. Table 7 also highlights the additional energy consumption, which compared to the energy input to the HPU ( $\sim 594~\text{GWh/a}$ ) corresponds to 16.7% and 4.9% for the CM and EEM solution, respectively.

#### 4.2. Heat supply curves for partial to full capture

Fig. 9 a shows how the cost of heat supply increases with steam demand from 1 to 24  $\varepsilon$ /t steam. Fig. 9 b and Fig. 9 c show the composition of the cost-minimized and energy-minimized heat supply, respectively. Both solutions exhibit changes from being almost entirely based on residual heat to increasing shares of existing capacity, and finally new capacity. For the minimum-cost solution (Fig. 9c), natural gas (class II, red field in Fig. 9 b) enters the mix in the range of 40–55 MW where the maximum HCN capacity is reached with increased demand. From  $\sim$  60 MW onwards, the HCN with MVR can cover most of the demand cost-efficiently. Natural gas combustion in existing boilers enters the mix again from  $\sim$  80 MW onwards with increasing shares. Note that beyond 160 MW, new natural gas boiler capacity is installed. For the solution with minimum external energy (Fig. 9 c), the transition from HCN to an HCN with MVR occurs at 40 MW, thus avoiding the use

Table 7
Key performance indicators of the cost-minimized (CM) and external-energy-minimized (EEM) solutions to achieve a heat supply of 52.56 MW for single-stack capture from the HPU. The optimization was either run including typical variations (process data) or on annually averaged values.

KPI	unit	CM solution		EEM solution	
		incl. variations	excl. variations	incl. variations	excl. variations
Specific cost (average steam cost)	€2018/ MWh	7.94	6.28	9.17	7.26
Specific emissions	t CO <sub>2</sub> / MWh	0.044	0.026	0.003	0.003
Break-even €/t CO <sub>2</sub> price			50	<del>-</del> 70	
Annual cost	k€/a	5887	4656	6794	5385
Extra emissions (scope 1 & 2)	kt CO <sub>2</sub> / a	19.56	11.75	1.36	1.36
Electricity consumed	GWh/a	22.3	24.2	28.9	28.9
Natural gas consumed	GWh/a	77.0	43.6	0	0

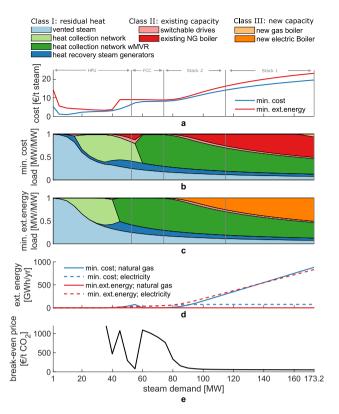


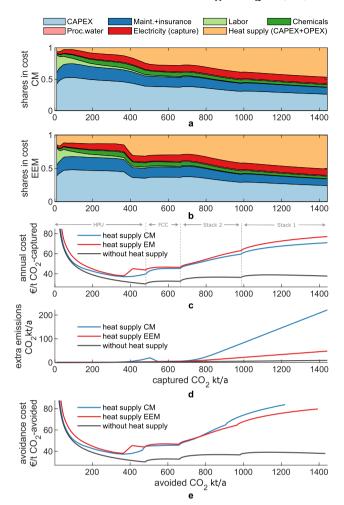
Fig. 9. Heat supply for partial to full  $CO_2$  capture for the studied refinery depending on the objective function CM (panel b) or EEM (panel c). Panel (a) shows the average cost of steam (sat. 131 °C). Panel (d) shows the external energy demand and panel (e) shows the break-even price of  $CO_2$  at which the costs for the two solutions are identical. The shaded areas in panel (b) and (c) indicate the classification of the heat sources. Notations: "NG", natural gas; "wMVR", with mechanical vapor recompression;

of natural gas, as compared to the cost-minimum solution. The use of natural gas is avoided entirely by the installation of an electric boiler when the capacity of the HCN MVR is depleted (from  $\sim 80$  MW onwards). Ranges of steam demand can be identified which would allow the freedom of extending the steam demand without running the risk of increasing system cost per tonne steam dramatically ( $\sim$ constant cost, e. g., 20–35 MW, 60–75, or >100 MW). Fig. 9 d shows the external energy demand and illustrates a similar consumption of external energy however of different character (high electricity use and high natural gas use for minimum energy and minimum cost solution, respectively, and viceversa). Fig. 9 e shows the break-even price of CO<sub>2</sub> at which the cost for both solutions become identical. High break-even prices indicate areas where the energy minimization solution has relatively high cost compared to the emissions saving potential.

#### 4.3. Site-level avoidance cost curve for CO2 capture

Fig. 10 compares the heat supply cost obtained via CM and EEM (Fig. 10 a and b, respectively) to cost elements of the capture plants capital and operational expenditures and shows the resulting capture cost (Fig. 10 c) and avoidance cost (Fig. 10 e). Also, the emissions from heat supply are compared to indirect emissions from electricity consumption in the capture plant (pumps, flue gas fan, air cooling) in Fig. 10d.

Fig. 10 c shows that reducing the capture rate in order to have heat supply predominantly based on Class I heat sources (residual heat) can lead to lower capture cost. This is partly due to lower heat supply cost, but also due to limited economy of scale effects (visualized by the black line in Fig. 10 c). Although the specific cost fall dramatically due to scale



**Fig. 10.** Impact of heat supply cost on capture cost (a-c), extra scope 1+2 emissions (d), and the avoidance cost (e) for  $CO_2$  capture at the studied refinery. The blue lines and panel (a) show the impacts for heat supply with minimum cost (CM), the red lines and panel(b) show the impacts for heat supply with minimum external energy consumption (EEM). The black lines show the impacts for the amine capture plant without heat supply. Note that the abscissa in panel (e) is different than in panel (a-d) and that the ordinate in panel (c) and panel (e) do not start from zero for illustration purposes.

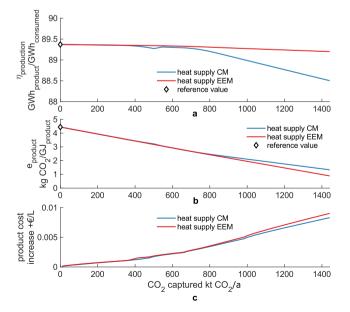
for the first stack, the cumulative specific investment for capture plants increases with the addition of the next stack in the merit order (one capture unit per stack). Avoidance cost as low as  $38~\rm fc/t~CO_2$ -avoided are reached in a range of  $340-400~\rm kt~CO_2/a$ , which corresponds to capture rate of  $70-83~\rm fc$  from HPU flue gases. Compared to this partial capture range, the avoidance cost is more than twice as high at  $80-84~\rm fc/t~CO_2$ -avoided when capturing 90% of the  $CO_2$  from the four major stacks. Fig.  $10~\rm a$  and b illustrate the increasing share of heat supply cost in annual cost with captured  $CO_2$ , which reaches  $\sim 50\%$  for all-stack capture for both CM and EEM solutions. Note the significant increase in extra emissions (scope 1+2) from  $\sim 700~\rm ktCO_2/a$  onwards which is due to increasing shares of Class II and Class III heat sources which require the import of external energy (cf. Fig. 9 c and d from  $\sim 80~\rm MW$  onwards).

External energy minimization leads to higher capture cost per tonne  $CO_2$  captured than cost minimization, as shown in Fig. 10 c. The maximum amount of avoided  $CO_2$  emissions is 1,394 kt  $CO_{2,avoided}/a$  and 1,220 kt  $CO_{2,avoided}/a$  for CM and EEM, respectively. This consequently affects the avoidance cost for  $CO_2$  capture at the site. Capture with heat supply using minimum external energy (electric boiler) has lower costs per tonne  $CO_2$  avoided for site capture rates > 770 kt/a than the solution that minimizes heat supply cost relying on natural gas.

The effect of heat supply on the KPIs is depicted in Fig. 11. The production efficiency falls slightly with increasing capture at first and falls more steeply from 700 kt CO<sub>2</sub>/a onwards. Depending on heat supply option (CM or EEM), the maximum loss in production efficiency is 0.17 – 0.87 percentage points. If the avoided emissions are related to the products, the production (refinery) emissions can be reduced from 4.44 kg CO<sub>2</sub>/GJ to 0.89–1.33 kg CO<sub>2</sub>/GJ, which corresponds to avoided refining emissions of 70–80 %. Note that fossil transport fuels emit  $\sim$  94 kg CO<sub>2eq</sub>/GJ [63]. The cost increase per litre of product increases with the site capture rate and is below < 0.01  $\epsilon$ /L (excluding liquefaction, CO<sub>2</sub> transport and storage).

#### 4.4. Sensitivity of findings with respect to techno-economic parameters

Fig. 12 shows the single-parameter sensitivity of steam cost and avoidance cost with respect to the selected parameters. The resulting steam cost is  $4-13 \notin /t$  steam and  $12-33 \notin /t$  steam for  $CO_2$  capture from the HPU and all stacks, respectively. The avoidance cost is 35 - 60 €/t CO<sub>2,avoided</sub> and 59 - 101 €/t CO<sub>2,avoided</sub> for capture from HPU and all stacks, respectively. Although the avoidance cost approach each other at low electricity and gas prices, it is clear from the figure that single-stack capture (HPU) achieves lower avoidance cost than all-stack capture for any of the individually varied parameters in their defined span of values. Of the tested parameters, energy prices and the discount rate (CAPEX) have the largest impact on heat supply cost and avoidance cost. Also, compared to capture from HPU, the all-stack capture scenario seems more sensitive to energy systems related parameters such as electricity price and emissions intensity of electricity and gas, and less sensitive to the chosen steam temperature and slightly less sensitive to the discount rate. Note that Fig. 12 also indicates changes in the default solution, i.e., when the cost-minimized solution starts using an electric boiler instead of natural gas or starts choosing the MVR option in the HCN (only relevant in the HPU case, since its already used by default for all stack capture). Further detailed findings are listed in Supplementary Materials S.6.



**Fig. 11.** Key performance indicators for the effect of CCS implementation on refinery operations: production efficiency (panel a), production emissions intensity (panel b) including scope 1 & 2 emissions, increase in production cost. Note that the ordinate in panel a does not start from zero.

#### 5. Discussion

#### 5.1. Variations in heat supply

The scenario analysis has highlighted the intermittent character that residual heat may have and the need to manage these intermittencies with other heat sources to supply a constant load of heat to the reboiler in capture plants. Representative hourly variations for an entire year have been considered, and thus the maximum installed capture and heat supply capacity (CAPEX) needed. Considering multi-year variations may reflect rare events better and thus would enhance the robustness of the suggested solution in handling such events. The impact of variations/ intermittency of available residual heat has been illustrated for singlestack capture (cf. Section 4.1): Cost and emissions increased by 26% and 66%, respectively. When Class I heat sources are less dominating, for instance for all-stack capture, the cost and emissions of heat supply would increase by up to 7% and 9%, respectively. It should be noted, that the uncertainty of the optimization of heat supply was not quantified here. Since the optimization is linear, a global optimum is achieved, thus the uncertainty is a function of the representiveness of the data, the assumptions made, and the linearization of cost functions (cf. Section S.4).

This work did not consider the dynamic operation of the system (including start-up and shut-down procedures of the capture plant), which could be a limitation if load-following heat generation and the steam transporting system are not capable of adapting to steep gradients in Class I heat sources. Thus, the premise of a constant heat load may not be practically feasible or, on the other hand, not needed if variations in heat supply can be handled by flexibility in the capture plant. Previous work [64] found that variations in reboiler duty at the scale of 1-3 h have limited effect on the effectiveness of the CO<sub>2</sub> capture. The magnitude of such resilience to variations is a function of the design, yet fundamentally caused by 1) the amine solvent circulating between stripper and absorber acting as a buffer and 2) a non-linear behaviour to heat load changes, i.e., an increase in heat load happens faster than a decrease of same magnitude (thermal inertia). Therefore, it may be preferable to allow the heat supply to the reboiler to vary instead of combusting natural gas to manage the variations in heat supply. A prerequisite for this is that the utilization of the capture plant (excessive CAPEX) does not drop significantly. For long-periods, seasonal variations, Eliasson et al. [19] find that exploiting residual heat only at the expense of larger equipment often not running at full capacity generally leads to higher avoidance cost than utilizing residual heat and external energy (Class II) at a constant load in smaller, fully utilized equipment. However, this depends on the energy prices, the degree of utilization and potential other uses for residual heat, for instance, district heating (causing the seasonal availability of residual heat to CCS), see [19].

#### 5.2. The value of residual heat and the crux of extra emissions

The results demonstrate that use of residual heat (Class I) - despite their partly intermittent character - is preferable (cost and emissions) to running existing (Class II) or new-installed boilers (Class III). If Class I heat sources are neglected for all-stack capture, the steam cost would dramatically increase to  $\sim 32~\text{€/t}$  or  $\sim 40~\text{€/t}$  steam for new natural gas boilers or electric boilers, respectively. The avoidance cost would increase to 112 and 132 €/t CO $_2$  avoided, respectively, implying a cost increase of 40–57%. Since these numbers account for economy of scale (see Supplementary Materials S.4.2.2), it is clear that the OPEX for external energy dominates. The required external energy supply would increase to 1,734 GWh/a and 1,494 GWh/a with respective emissions of 411 kt CO $_2$ /a and 79 kt CO $_2$ /a for natural gas boilers and electric boilers, respectively.

The large emissions from unmitigated fossil fuel use assumed here impact the avoidance cost significantly (cf. Fig. 10). The decision to not capture these emissions was made, 1), to keep calculations simple and

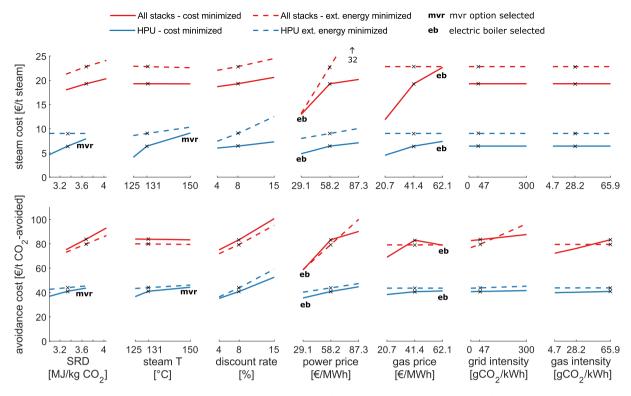


Fig. 12. Sensitivity of heat supply cost (top panel) and avoidance cost (bottom panel) for single-stack capture from HPU flue gas (blue) and all-stack capture (red) towards specific reboiler demand (SRD), steam temperature (sat.), discount rate, electricity price, gas price, electricity CO<sub>2</sub> intensity and gas CO<sub>2</sub> intensity. The markers (x) show the default values for which the mix of heat sources is identical to Fig. 9. The notations "mvr" and "eb" indicate when the default composition changes to include mechanical vapor recompression and an electric boiler, respectively. The dashed lines represent the energy-minimized solutions, the full lines the cost-minimized solutions of the HSCM.

transparent, and, 2), to show the impact of fossil fuel use. However, if these extra direct emissions were to be captured with 90% efficiency in the case of all-stack capture, the capture cost would increase by 4.6 % to 74  $\mbox{\'e}/\mbox{t}$  CO<sub>2</sub>. The amount of unmitigated extra emissions would decrease by 69% to 66.5 kt CO<sub>2</sub>/a, since not all direct emissions are captured and indirect emissions in the NG supply (9.7 gCO<sub>2</sub>/MJ) are unmitigated. The avoidance cost would fall by 7.1% to 77.8  $\mbox{\'e}/\mbox{t}$  CO<sub>2,avoided</sub>. In comparison, supplying heat for all-stack capture with an electric boiler has slightly higher avoidance costs (79.7  $\mbox{\'e}/\mbox{t}$  CO<sub>2,avoided</sub>), yet has lower extra emissions (48 kt CO<sub>2</sub>/a). From this exercise, we conclude that a rigorous consideration of indirect emissions from external energy is necessary and that an electric boiler can be an economically viable option for heat supply if the grid CO<sub>2</sub> intensity is low enough and the electricity prices are reasonable. This is in line with conclusions by Roussanaly et al. [30].

#### 5.3. The role of CCS in future bio-refinery configurations

As shown in Section 4.3, the impact of CCS on cost and emissions reduction of the refinery's end-products is relatively small. However, refineries that transition to bio-refineries will have a lower share of fossil emissions in the end-product, which could lead to a higher importance of refining emissions – if these continue to have a fossil origin. Note that higher shares of biogenic feed require a higher hydrogen input to reach the O/C and H/C-ratios needed in the products [65], which could increase direct refining emissions if steam reforming/gasification of hydrocarbons is applied. If these hydrocarbons are of biogenic origin, the CCS implementation could even lead to negative CO<sub>2</sub> emissions [66].

#### 5.4. The usefulness of energy supply cost curves

The usefulness of ECC is illustrated for heat supply in the case study of the Swedish refinery. The cost of heat supply increases with  $CO_2$ 

capture rate and the abatement costs are significantly affected once the residual heat is recovered. One strength of the ECC approach presented here is that it covers the entire span from partial to full capture at site level. For instance, the identification of lowest cost for 70-83% of capture from the HPU (cf. Fig. 10 e), was only possible by resolving the capture and heat supply cost for various capture rates from individual stacks. Similarly, the approach helped identify areas of flat cost response (cf. Fig. 10 e, between 500 and 650 kt CO<sub>2,avoided</sub>/a). Considering entire stacks as marginal increment, as often is done in marginal abatement cost curves [9,13], does not capture the detail of insights presented in this work. Note also, that the cost for relocating existing equipment or interconnecting cost (ducting of flue gases/solvent) due to contraints on available plot area on site were not included here. These are, however, discussed in the IEAGHG report [14], wherein the interconnecting cost in refineries were found to make up 10-20% of the total cost for retrofitting CO<sub>2</sub> capture and conditioning cost to refinery sites.

The abatement cost curves presented in this work do not include liquefaction, interim on-site storage, transport and storage of CO $_2$ . Including rough estimations of liquefaction according to [34] and transport and storage to the Utsira formation (North Sea) by ship according to [67] would lead to full-chain capture cost of  $\sim 78~\rm fe/t~CO_{2,captured}$  and  $\sim 102~\rm fe/t~CO_{2,captured}$ , for single-stack capture from HPU or all-stack capture, respectively (see Supplementary Materials S.7). The assessed heat supply cost would comprise 13-33%, which shows that differences in heat supply cost with the degree of CCS implementation at the refinery site have significant implications for the full-chain cost as well. This is in line with FEED study reports of the Norwegian Longship project which report utility cost in the range of 33% [68].

#### 6. Conclusions

In this work, we have introduced the concept of energy supply cost

curves (ECC) for CCS, which identifies cost of energy supply as a function of the level of CCS implementation and the existing site energy system. For this purpose, a systematic techno-economic analysis using multi-period mixed integer linear programming was conducted to find a cost or energy minimal mix of heat sources. The residual heat of the refinery process and the capacity of generating steam in existing equipment was explicitly considered, as well as the characteristic temporal variations of these heat sources. We illustrated the concept for heat supply for amine solvent regeneration in a case study of a Swedish refinery, from which we conclude the following:

- Intermittency of residual heat needs to be managed by flexible load-following heat sources, such as gas or electric boilers or sufficiently large heat collection networks. The inclusion of temporal variations instead of using annually averaged values in the analysis led to higher cost of 7–26% and emissions of 9–66%, depending on the share of the intermittent heat sources in the mix.
- Significant shares of the site emissions ( $\sim$ 40%) can be captured utilizing residual heat alone, and  $\sim$  78% of all site emissions could be captured in combination with existing boiler capacity. This energy and capital cost-efficient use of the existing site energy system reduced heat supply cost, which were estimated to be in the range of 1–33  $\rm €/t$  steam. This large span in heat supply cost significantly affects the cost of CO<sub>2</sub> capture (excl. liquefaction, transport and storage) which were estimated to 35 60  $\rm €/t$  CO<sub>2,avoided</sub> and 59 101  $\rm €/t$  CO<sub>2,avoided</sub> for 90% single-stack capture from the hydrogen production unit and 90% from all four major stacks, respectively. For the examined site and its site energy system, partial capture from a single stack is economically more viable than full capture, and thus presents a starting point for carbon capture at the site.

We have demonstrated that the inclusion of ECC into abatement cost curves at site level can generate valuable insights for a low-cost CCS implementation at industrial sites. These may help to plan roadmaps that start off with low-cost partial capture and thereafter move on to full capture or to the combination with other mitigation measures, to enable net-zero emissions from an industry site.

#### CRediT authorship contribution statement

Maximilian Biermann: Conceptualization, Methodology, Software, Data curation, Visualization, Validation, Writing – original draft. Christian Langner: Methodology, Software, Writing – original draft, Writing – review & editing. Simon Roussanaly: Resources, Writing – review & editing. Fredrik Normann: Conceptualization, Supervision, Writing – review & editing. Simon Harvey: Funding acquisition, Project administration, Supervision, Writing – review & editing.

#### **Declaration of Competing Interest**

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

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#### Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apenergy.2022.119273.

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