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# Production of marine biofuels from hydrothermal liquefaction of sewage sludge. Preliminary techno-economic analysis and life-cycle GHG emissions assessment of Dutch case study

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ABSTRACT

The aim of this paper is to evaluate the costs and GHG emissions of advanced biofuels production through hydrothermal liquefaction (HTL) of sewage sludge in The Netherlands targeting the marine fuels market. The process evaluated consists of a distributed configuration of regional HTL plants co-located with wastewater treatment plants, with centralized hydrotreating co-located with an existing refinery at the Port of Rotterdam. The process is simulated in ASPEN + based on published experimental data and the mass and energy balances are used as input for techno-economic and environmental evaluation. Lifecycle GHG emissions of the HTL and hydrotreating processes are estimated using consequential modelling principles and background data from the Ecoinvent database and compared with the business-as-usual scenario of sludge mono-incineration and fossil marine fuels production. The results indicate that the HTL + hydrotreating configuration has potential to deliver on-spec marine biofuels at a minimum fuel selling price between 410 and 1300 EUR/t, being at least 3 times more beneficial compared to the business-as-usual scenario from a GHG emissions perspective. Future work is recommended to optimize the size and location of the HTL plants in order to decrease capital costs and to address uncertainties regarding the sludge gate fee and the costs associated with the aqueous and solid by-products treatment. The results indicate the potential of such configuration in locations with relatively high population density and good transport infrastructure. This can be the case of port areas around the North Sea with access to offshore renewable electricity for hydrogen production, where drop-in marine biofuels are expected to play a role with the increasing share of renewables in the marine fuels mix.

## Introduction

Increasing circularity in the management and recovery of resources has been highlighted in the European Green Deal as essential to achieve the climate targets and to assure a sustainable economic growth. In this context, it is key to develop technologies for waste valorization that can be integrated in a systemic solution for the energy transition.

Sewage sludge is a type of urban waste produced in large quantities as byproduct in the wastewater treatment plants. It contains organic and inorganic compounds separated from the aqueous influent after undergoing different types of treatment (i.e. primary and secondary). Even though sludge composes mainly water (70–80 % based on mass), its organic fraction is a valuable source of carbon and nutrients that can be further recovered and utilized. In 2016, approximately 7 million dry tonnes of sludge were produced in Europe and disposed mainly through agricultural use and incineration with 34 % and 33 % of the total respectively [1]. Despite of allowing for some degree of energy/nutrients recovery in the form of heat and fertilizers, current disposal methods face challenges due to strict emission limits and high disposal costs in incineration plants, and regulatory limitations for sludge disposal in the fields, with increasing concerns regarding presence of pathogenic compounds, pharmaceutical and personal products (PPCP), microplastics, heavy metals etc.

Sewage sludge treatment through hydrothermal liquefaction (HTL) is one of the alternatives to the existing methods that has gained momentum due to the efficient conversion of the sludge's organic fraction into biocrude, a product that can be further processed into advanced transportation biofuels. The aqueous and solid by-products obtained in

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the process contain most of the nitrogen and phosphorous in the sludge, and additional treatment can be integrated for their recovery and use as fertilizers. The hydrothermal conditions used in the HTL process, in the range of temperatures between 250 and 450 °C and pressures between 100 and 350 bar approximately, have resulted in the effective degradation of different types of pharmaceuticals, biocides, and other bioactive compounds [2,3].

Sewage sludge valorization through HTL has been documented in literature with several studies that investigate the impact of different process conditions in the products yields and characteristics [4–8], as well as the techno-economic feasibility that has resulted in an estimated minimum fuel selling price (MFSP) between 0.8 and 1.4 USD/L approximately [9,10]. Furthermore, the announcement of demonstration projects and cooperation agreements for the development of the technology in Norway, Canada, and Australia [11,12] could lead the way for future HTL projects in an European context, where the techno-economic and regulatory feasibility has not been widely approached in existing literature. In the field of hydrothermal treatment, most of the studies addressing costs of sewage sludge valorization in Europe have been focused on hydrothermal carbonization [13–15], while much less attention has been given to HTL [16]. In a recent publication, Castro et al. evaluated the integration of HTL and catalytic hydrothermal gasification (CHG) in the operation of wastewater treatment plants showing different configurations that resulted in a reduced wastewater treatment cost and environmental impact [17]. Additionally, there is lack of studies that evaluate sludge valorization through HTL from a systemic approach, looking at regional or national feedstock availabilities and biocrude production potentials, with only a recent contribution from Seiple et al. in the US [18].In terms of market uptake, sustainable advanced biofuels are considered key in the energy transition to enable rapid emission reductions in sectors such as maritime, as its implementation does not require significant changes in the existing infrastructure. Particularly in the North Sea, decarbonization of the maritime transportation is key due to its major contribution to the region's economy and the high environmental impact caused by its dependency on fossil fuels. According to the International Environmental Agency, current global marine fuels consumption is estimated to be around 330 million tonnes/year, and demand is predicted to double by 2030 as global trade increases. Even though the sector is not currently subject to obligation in the Renewable Energy Directive – Recast to 2030 (REDII), the Fuel EU Maritime initiative launched in 2021 aims to increase the use of sustainable marine fuels in European shipping and ports, which account for about 11 % of all European $CO_2$ emissions from transport and 3 to 4 % of total EU $CO_2$ emissions [19]. Furthermore, regulations set by the International Maritime Organization (IMO) limit the sulfur content in marine fuels, which implies that about 70 % of the fuels currently used by the sector needs to be modified or changed [20].

Given the relevance of the topic and the research gaps encountered, the purpose of this study is to evaluate from a systemic approach the deployment of the HTL technology as an alternative to current sludge disposal methods, and to assess the system potential to deliver sustainable advanced biofuels with focus on the marine sector. This study selects The Netherlands as case study due to its developed transport infrastructure and dense population, and the presence of one of the main ports in Europe, the Port of Rotterdam. Currently, about 70–90 % of the sludge produced in The Netherlands is incinerated at a substantial gate fee that can be estimated in the order of 100 EUR/t [21], giving opportunity for the deployment of other alternatives that can provide solutions in other sectors of the energy system.

From a techno-economic perspective, the objective of the paper is to evaluate a configuration of HTL and biocrude upgrading steps integrated with existing industries, namely wastewater treatment plants (WWTPs) and fossil refineries, and to estimate the cost of sludge treatment and the final MFSP of the marine biofuels having The Netherlands as case study. Particular emphasis is given to the drop-in potential of the HTL biocrude relative to current marine fuel specifications using in-house experimental data. Based on the results, the aim is to discuss the opportunities and main barriers for the deployment of the HTL technology for sewage sludge valorization from a techno-economic and environmental perspective, using the Dutch case as an example that can potentially be extrapolated to other locations.

The structure of the paper comprises a methodology section followed by results and discussions and finally conclusions. The first section presents the considerations regarding feedstock availability and the detailed process configuration of the HTL and upgrading steps. Next, the results are discussed in terms of: 1) sewage sludge regional distribution; 2) mass and energy balances, drop-in potential and MFSP of the marine biofuels, and 4) LCA assessment in comparison with current disposal via incineration.

## Process description and methodology

### *Feedstock availability*

The assessment of feedstock availability is conducted based on publicly available statistics of sewage sludge production and disposal in The Netherlands in 2018 reported by *Statistics Netherlands* (CBS) [22], being this the last year reported at the moment of the study. The data available is aggregated in national and regional levels and it provides the total production and its destination by activity (disposal method). The sludge disposed via mono-incineration is the targeted feedstock for the HTL process. Data available on dry matter and ash content is used to estimate the organic, inorganic and aqueous fractions going through the process.

In the Netherlands there are more than 320 WWTPs with capacities between 2,300 and 765,000 person-equivalent (p.e) [23], equivalent to 0.46–153 ML/d approximately. Since sludge production per WWTP is unknown, the WWTP influent capacities reported in [23] are used as an indication of their relative sludge production. It is estimated that about 45 % of the total sludge is produced in relatively small or medium size plants with capacity below 20 ML/d that account for 85 % of the total number of plants (Fig. 1)[1]. Following a top-down approach, a regional

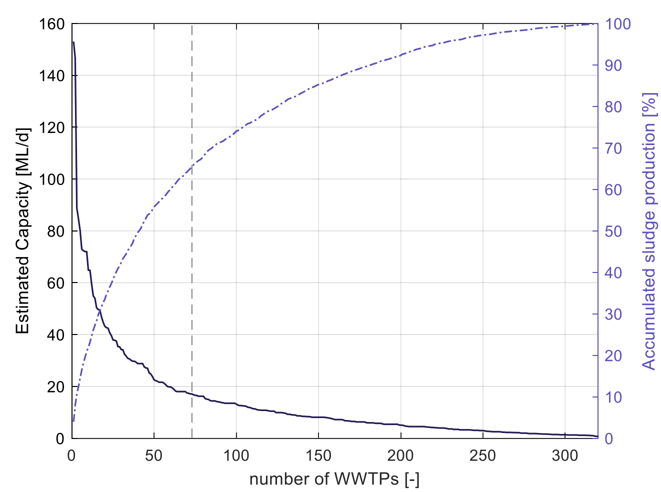


**Fig. 1.** Treatment capacity per WWTP in The Netherlands and accumulated sludge production.

[1] Accumulated sludge production estimated based on the reported COD of the influent wastewater per WWTP [23] following the assumption reported in [18]: 67 % of the COD is obtained in the sludge after primary and secondary treatment and is converted to energy basis based on a factor of 13.9 kJ/g COD (reported as the maximum attainable from methanogenic wastewater treatment [57])

scheme of HTL plants that gathers sludge from nearby plants is evaluated to harness the full theoretical potential from the smaller WWTPs.

*HTL and upgrading process*

*Distributed HTL plants with centralized upgrading*

This study comprises a configuration of regional HTL plants co-located with existing wastewater treatment plants (WWTPs), or in proximity of a cluster of WWTPs, and a centralized hydrotreating unit (UPG) co-located with existing fossil refineries at the Port of Rotterdam (Fig. 2).

WWTPs and refineries are the selected sites for co-location, taking advantage of existing infrastructure for utilities and hydrogen supply and due to the strategic location for delivering marine fuels close to final fuel users. It is assumed that the aqueous by-product from the HTL plant can be recirculated to the host WWTP influent or other entry points of the WWTP assuming that there are no inhibition or similar negative impacts in the WWTP operation due to the high dilution factor, and the HTL combustible gas is evaluated for internal heat supply. The HTL solid residue (mineral product) is a main by-product from the process and is discussed in the context of phosphorous recovery. The HTL biocrude is sent for upgrading to the centralized location, where biocrude hydrogenation is performed in a dedicated unit (no co-refining with fossil crude is assumed) in connection with the refinery mainly for $H_2$ supply and $H_2$ recovery from the hydrotreater off-gas (described in Section 2.2.2). The indirect emissions associated with the exchanges with the WWTP and the refinery are accounted for in the environmental assessment, described in Section 2.3.

The regional aggregated data of sludge available per year is used to estimate the number of HTL plants in each region assuming a fixed capacity of 100 dry tonnes per day (t/d) and 8,000 operational hours per year. This value is used as first approach in this study in absence of higher resolution data regarding the sludge production per WWTP, and based on the value of 110 dry t/d used in [9]. The largest WWTPs in the regions are the selected locations for the HTL plants. This information is used to estimate the size of the upgrading unit and road transportation costs within regions and to the Port of Rotterdam.

*Modelling approach*

The system modeled regarding mass and energy balances consists of the HTL and hydrotreating units (inputs and outputs of HTL and UPG boxes in Fig. 1). A generic modeling approach for the HTL and hydrotreating processes applicable to different feedstock is applied in this study for sewage sludge in the context of The Netherlands. The co-location assumption does not have an influence in the methodologies used. For HTL the method is applicable to different feedstock and in this case is evaluated for final sludge treatment. Likewise, the methodology used in the hydrotreatment is not affected by co-location in a refinery under the assumption that a dedicated unit is used for the biocrude without co-refining with fossil crude.

The HTL and upgrading processes are modeled in Aspen Plus® V9 (ASPEN + ) based on state-of-the-art and in-house experimental data on sewage sludge using the Soave-Redlich Kwong (SRK) property method, which provides reliable results in the critical region and is recommended for hydrocarbons applications.

The following sections present the approach and main considerations in each step. The methodology used for the modelling of the HTL and the hydrotreating units has been described in detail in previous publications based on woody biomass [24,25], and is extended to sewage sludge in the present study. Overall, the global mass balances on the HTL and hydrotreating processes are based exclusively on experimental yields reported in literature, and the experimental characterization of the biomass and the different products is used to establish mass and energy balances.

The feedstock characterization data used in this study corresponds to a sample from a Dutch WWTP analyzed under the NGRF project [26] (Table 1). Given that all the WWTPs reported in [23] and accounted in this study include secondary treatment, only one HTL plant is modeled under the assumption that there are not significant variations in the quality and properties of the sludge feedstock across the country. Data on feedstock variations is unknown in this case study preventing a more detailed assessment; however, this assumption is considered reasonable for the purpose of preliminary techno-economic evaluation due to the relatively small variations documented in the energy content of HTL biocrudes across waste biomasses, and the lower influence of the feedstock composition variability in the economic evaluation relative to other cost parameters with larger influence such as feed moisture and capital investment [27,28].

Hydrothermal liquefaction of sewage sludge

Sewage sludge entering the process with a dry matter content of 25 wt% is heated and fed to the HTL unit. Drying of the sludge is not necessary before the HTL process since the dry matter is already adjusted to this level for current disposal methods [22]. The sludge is modeled as a non-conventional compound with modified settings of the HCOALGEN enthalpy model, following the procedure described in detail in [24]. Proximate and ultimate analysis are used as input data and the enthalpy of formation is estimated from the user-input experimental high heating value (HHV) (Table 1). The HTL reactor is modeled with the User 2 model linked to Excel® specifying the products yields and compositions (Table 2). The yields are specified in agreement with values in published literature of HTL for sewage sludge [3,6,27,29].

The biocrude is modeled using the model compounds approach, adjusting its composition by multi-objective optimization to match the experimental data shown in Table 1. The procedure is described in detail in [24] and the composition used can be consulted in the Supplementary material. The gaseous product is modeled based on the composition

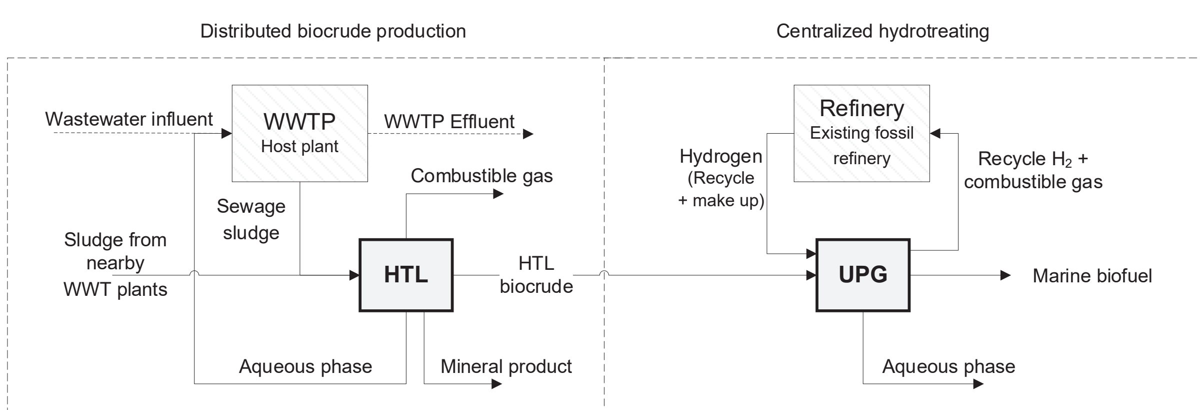


**Fig. 2.** Scheme of distributed configuration of regional HTL plants with centralized hydrotreating (UPG).

**Table 1**
Elemental composition of sewage sludge, HTL products and hydrotreated biocrude (dry basis).

| | C [wt.%] | H [wt.%] | O [wt.%] | N [wt.%] | Ash [wt.%] | Fixed carbon [wt.%] | Volatile matter [wt.%] | HHV [MJ/kg] |
|---|---|---|---|---|---|---|---|---|
| Sewage sludge | 38.2 | 6.6 | 23.9 | 6.4 | 24.9 | 6.2 | 68.9 | 18.2 |
| HTL biocrude [8] | 74.5 | 10.6 | 11.0 | 3.9 | NR | NR | NR | 37.4 |
| Hydrotreated biocrude [8] | 85.3 | 13.8 | 0.0 | 0.9 | NR | NR | NR | 46.1 |
| HTL mineral product [6] | 15.8 | 0.7 | 4.3 | 1.5 | 77.6 | NR | NR | NR |

*NR: Not reported.

**Table 2**
Main input parameters of HTL and hydrotreater units in Aspen Plus®.

| Unit | Process conditions | Modeling parameters |
|---|---|---|
| HTL reactor | T = 350 °C, P = 200 bar. | Gas yield ($Y_{g/ss}$ = 0.15 kg/kg) and composition (Table 3)<br>Biocrude yield ($Y_{bc/ss}$ = 0.40 kg/kg) and composition<br>Solids yield ($Y_{s/ss}$ = 0.10 kg/kg) and elemental composition (Table 1) |
| Hydrotreater | T = 400 °C, P = 120 bar | Gas yield ($Y_{g/bc}$ = 0.085 kg/kg) and composition (normalized excluding $H_2$ in Table 3)<br>Biocrude yield, $Y_{bc/bc}$ = 0.841 kg/kg and elemental composition (Table 1)<br>Hydrogen consumption = 0.04 g $H_2$/g HTL biocrude |

*Subindices g = gas, bc = biocrude, s = solid, ss = sludge (daf = dry ash-free basis).

presented in Table 3 and the mineral product is modeled as non-conventional solid as indicated for the sludge based on the proximate and ultimate analysis in Table 1. The composition of the inorganic fraction in the mineral product is not modeled in ASPEN+; however, the phosphorous (P) content is of main interest for the overall mass balance and environmental assessment and it is estimated based on a 95 wt% P recovery relative to the sludge [6]. The P in the sewage sludge reported by CBS in [22] (reference year 2018) is used for the estimation.

Regarding the aqueous phase there is no sufficient experimental data available to fully describe its composition and therefore it is adjusted to close the overall carbon and nitrogen balance assuming that mainly small organic acids and nitrogenates are present, based on the reported composition by Madsen et al. [30]. The products from the HTL reactor are cooled down and decompressed to ambient conditions to be separated. The HTL off-gas is assumed to be combusted for heating supply, so its energy potential is included in the energy balance to reduce the external heating. The utilities requirement (electricity, heating and cooling) is obtained from the simulation results and is used to evaluate heat integration between the process streams based on pinch analysis (minimum terminal temperature difference of 20 °C is selected in agreement with typical values between 10 and 20 °C [31]).

<u>Upgrading of HTL biocrude</u>

The HTL biocrude is conditioned to the temperature and pressure to be upgraded via hydrotreatment (e.g., hydrogenation, hydrodeoxygenation and hydrodenitrogenation). This process uses hydrogen to remove residual oxygen molecules and other heteroatoms transferred from the feedstock. The hydrotreater is modelled as a single step with the User 2 model linked to Excel® based on reported product yields (Table 2).

Available in-house experimental results (hydrotreated biocrude yield, true-boiling point (TBP) curve, density and sulfur content) are used to model the hydrotreated biocrude and predict additional properties based on the petro-characterization tools available in Aspen Plus®. The difference in the oxygen and nitrogen content between the HTL and the hydrotreated biocrude is used to estimate water production via hydrodeoxygenation, and ammonia via hydrodenitrogenation.

For the hydrotreating process $H_2$ is required in excess (0.4 g/g input HTL biocrude) but the consumption is only the 10 % of the input (0.04 g/g oil) which is in the order reported in [8]. The remaining $H_2$ is the main component in the off-gas from the hydrotreater alongside $CO_2$, CO and hydrocarbons, produced in minor quantities during the hydrotreating process, and its recovery from the gas is necessary for the economy of the process. Since the hydrotreater is to be co-located with an existing refinery with infrastructure for $H_2$ purification, it is assumed that 98 % of the $H_2$ can be recovered via pressure swing adsorption (PSA) [32], and that make-up $H_2$ produced from electrolysis (78 % efficiency [33]) is fed at the conditions required to compensate for the $H_2$ consumption and losses in the recirculation loop.

**Table 3**
Composition of HTL and hydrotreater effluent gases.

| | HTL gas [mol. %] | Hydrotreater gas [vol. %] [8] |
|---|---|---|
| $H_2$ | 1.06 | 85.9 |
| $N_2$ | 0.53 | 0.00 |
| $CO_2$ | 89.84 | 2.50 |
| CO | 1.03 | 0.30 |
| Methane | 2.68 | 6.00 |
| $C_2$ | 1.16 | 3.33 |
| $C_{3+}$ | 3.07 | 2.00 |

*Drop-in potential of hydrotreated product*

Available in-house experimental work on biocrude hydrotreatment (upgraded oil yield, TBP curve, density and sulfur content) is used to model the fuel using petro-chemical tools in ASPEN + to predict additional properties. The drop-in potential is preliminary assessed by comparing these properties with the ISO marine fuel specifications [20] in the distillate range (marine gas oil (DMA/Z) or marine diesel oil (DMB)) and residual range (RMG 180 and 380). The properties evaluated (viscosity, density, flash point and sulfur content) are only few of the complete ISO specifications but are presented as an indication of the drop-in potential based on the modelling results, since a full assessment is out of the scope of this analysis.

*Life-cycle GHG emissions assessment relative to current sludge incineration and marine fossil fuels*

The purpose of the assessment is to evaluate the environmental performance of the conversion of sludge to fuel as an alternative treatment, compared to the current practice. Because the sludge is a waste, it is inherently constrained in supply so an increased demand for sludge-derived fuel will therefore not lead to increased production of the sludge-feedstock. Henceforth, the examined function is a waste treatment of sludge.

The functional unit of the assessment is treatment of 1 kg sludge dry weight, and two reference flows are examined. The first is the 'business as usual' (BAU) at the sludge treatment plant NV Slib Processing Noord-Brabant (SNB), which is described in detail in [34,35]. The second reference flow is the treatment process described in this article, plus combustion of the produced hydrotreated biocrude, minus the avoided production and combustion of the fossil analogue. In this way, the comparison includes a well to wheel assessment keeping the comparability between the BAU and the HTL technology scenario.

The modelling is done with consequential modelling principles

[36,37], using the consequential database Ecoinvent 3.6 implemented in SimaPro, modelling the background system. The results are presented in $CO_2$ equivalents ($CO_2e$) based on the IPCC 2013 GWP 100a method. Table 4 summarizes the scenarios evaluated and the main assumptions considered.

The BAU treatment is a mono-combustion process at SNB with utilization of byproducts and avoiding the corresponding extraction and production of phosphate rock, sand, quicklime and gypsum mineral. Special for the BAU scenario is that part of the flue gas, through a simple scrubber, is reutilized in a neighboring industry producing precipitated calcium carbonate (PCC), which in [34,35] is modelled as avoided carbon dioxide from natural gas. In the present assessment the SNB foreground system is remodeled with consequential principles and connected to the consequential background database, securing comparability between the reference flows. Details can be found in the supplementary material and data that can be imported into LCA-software can be sent upon request[2].

The alternative treatment through the HTL-hydrotreating pathway is modeled using the principles and transport distances as described in Section 2.2. In the assessment, standard energy consumption is assumed for $H_2$ re-pressurization, and a high efficiency of the PSA process and electrolysis to supply $H_2$ make-up as previously described. The most likely situation, in an implementation-scenario of the HTL pathway, is that electrolysis for hydrogen production is so widespread that the co-production of oxygen will exceed marked needs, implying no avoided production of oxygen. To maintain comparability with the basis scenario, waste streams of aqueous and mineral product from the HTL process are assumed being processed in the same way as in the BAU scenario, modeling the HTL mineral product as avoided phosphate rock, and aqueous products treated in a standard WWTP. In all scenarios, including BAU, the electricity supplies are modelled as the marginal Dutch supply as defined in Ecoinvent, and the efficiency of the electrolyser delivering the hydrogen make-up is defined to 78 % [33]. The impact of the avoided products from the HTL-hydrotreating pathway in the emissions is assessed in four different scenarios presented in Table 4. The most pessimistic scenario corresponds to the less integrated process where there are no other avoided products than the marine fossil fuel and the $H_2$ recovery efficiency for upgrading is relatively low (given the high $H_2$ purity of the PSA feed). The assumption of avoided phosphate rock is tested in the conservative scenario and the integration of excess heat from the process is tested in the BAU comparable scenario. The best case assumes the integration of all the different process streams. This evaluation allows to assess the impact of the main products in the GHG emissions separately.

*Cost estimation*

The parameters used in the cost estimation are summarized in Table 5. Regarding the capital costs, the total installed cost of the HTL and hydrotreating plants are adapted from the reported in [9,38]. Given the co-location approach, the investment costs related to the HTL aqueous phase treatment and the hydrogen plant are excluded from the original source. The original values reported in USD are scaled to the plant capacities of this study, using a scaling factor of 0.6 and converted to $EUR_{2018}$ using the Chemical Engineering Plant Cost Index and an average exchange rate of 0.85 EUR/USD in 2018. The total capital investment (TCI) and fixed operational costs are estimated using the Standardized Cost Estimation for New Technologies method (SCENT) [39], including the CAPEX and OPEX reductions reported by de Jong [40] due to co-location benefits.

The variable operational costs are shown in Table 5 for the base case, and minimum and maximum values are evaluated using the indicated percentage change when no other data is available. A sludge tipping fee of 100 EUR/t in the base case is in agreement with the disposal costs reported in the EU for mono-incineration [41,42]. The cost of hydrogen includes only the $H_2$ make-up and the price is evaluated based on the levelized cost of hydrogen reported in the Dutch market for electrolysis [43]. The impact of the hydrogen source in the cost is tested separately with the price of blue $H_2$ (produced from natural gas though SMR coupled with carbon capture and storage (SMR-blue)) reported in the same reference [43]. This scenario is considered interesting from an economic point of view in the short-medium term, however it is not tested in the environmental assessment as in the future the production of green $H_2$ is foreseen to be significantly cleaner due to lower electricity emission factor for The Netherlands based on the Simapro database (Table 4). The cost of heating is estimated based on the price of natural gas and assuming a boiler efficiency of 80 %. The cost of cooling provided by cooling water is estimated as 1 % of the cost of power, since the principal operating cost associated with its provision is the cost of power to drive the cooling tower fans and cooling water circulation pumps [44]. Operating labour costs are estimated based on an average EU hourly labour cost of 31.40 EUR/h [45] and the operating labour requirements indicated in literature for solids-fluid processing in a continuous operation (2 operators per shift and 5 shifts per week are assumed) [46].

Based on the capital and operational expenses obtained a net present value (NPV) analysis is performed to estimate the MFSP of the hydrotreated biocrude based on a 10 % discount rate, straight line depreciation and a project lifetime of 25 years. Monte Carlo simulations were performed with a random variation of the cost parameters in Table 5 in order to estimate the MFSP range expected of the HTL and hydrotreated biocrudes.

## Results and discussion

*Feedstock availability*

In 2018 the total production of sewage sludge in The Netherlands was 1.25 million tonnes (Mt), out of which 880 thousand tonnes (kt) were incinerated mainly in the west and south regions (Fig. 3). The wet feedstock from the WWTPs has in average a dry matter content of 24.5 wt%, out of which 29 % in average is reported as ash, leaving approximately 153 kt/y of organic matter to be valorized through HTL (Table 6).

*HTL and upgrading process*

*Distributed configuration of HTL plants with centralized upgrading*

The overall results of the HTL and hydrotreated biocrude potentials are summarized in Table 6. Based on the national and regional quantities reported, it is estimated that in total about six HTL plants of 100 dry t/d capacity each, (equivalent to 133 kt/y approximately including water), would be needed to replace the current disposal via mono-incineration. There is no certainty about the number of mono-incinerators currently operating in The Netherlands, however, the mono-incinerator located in Moerdijk, south-west of the country, processed 434.8 kt in 2019 [21], which corresponds to about 50 % of the total sludge incinerated. The size of the regional HTL plants is about 30 % of this size, so the proposed scheme is considered realistic since already large amounts of sludge are being transported across regions. There are no plants allocated in the east region since the sludge available is less than half of the model plant capacity. By excluding it, only 2 % of the targeted sludge is left out and the total HTL biocrude production potential is then rounded to 60 kt/y.

The tentative location of the plants and the average distances to cover within regions and to the upgrading facility are presented in Table 7, including the capacities of the hosts WWTPs that are included as reference for the mass and energy balances presented in the next section. The distributed configuration of HTL regional plants and centralized

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**Table 4**
Main assumptions in carbon footprint analysis of sludge mono-incineration (BAU) and HTL biorefinery scenarios.

| | Avoided fossil marine fuel | CCU of flue gas | Avoided phosphate rock (30%) /other products | Avoided fossil heat | $H_2$ recovery efficiency (PSA) |
|---|---|---|---|---|---|
| BAU | – | Yes: ~20% | Yes/Yes | No | – |
| HTL + UPG (worst case) | Yes | No | No/No | No | Low (90 %) |
| HTL + UPG (conservative) | Yes | No | Yes/No | No | Low (90 %) |
| HTL + UPG (best case – BAU-comparable) | Yes | No | Yes/No | Yes (30%) | High (98 %) |
| HTL + UPG (best case – scalable) | Yes | Yes: (as in BAU) | Yes/No | Yes (30%) | High (98 %) |

*NL Emission from marginal electricity consumption based on ecoinvent 3 (implementation in Simapro): [Electricity, medium voltage {NL}| market for | Conseq, U].

**Table 5**
Parameters for cost estimation.

| | Cost component | Unit | Base | Min | Max |
|---|---|---|---|---|---|
| CAPEX | Total installed cost of HTL plant | million $EUR_{2018}$ | 19.20 | 14.70 | 21.60 |
| | Total installed cost of UPG plant | million $EUR_{2018}$ | 17.40 | 17.40 | 29.40 |
| Variable Operational costs | Sludge credit (incineration) [21] | EUR/t | 100 | −20% | 0% |
| | Final solids disposal [47] | EUR/t | 133 | 0% | +100% |
| | Aqueous phase treatment [48] | EUR/$m^3$ | 4.26 | 0% | +100% |
| | Transport cost road [40] | EUR/t/km | 0.16 | −20% | +20% |
| | Loading and unloading [40] | EUR/t | 1.39 | 0% | +20% |
| | Electricity [43] | EUR/MWh | 47.00 | 40.00 | 60.00 |
| | Hydrogen from electrolysis [43] | EUR/kg | 2.80 | 2.50 | 3.70 |
| | Hydrogen from SMR with CCS [43] | EUR/kg | 2.00 | 1.60 | 2.60 |
| | Heating | EUR/MWh | 50.0 | 25.00 | 50.00 |
| | Cooling water | EUR/MWh | 0.47 | 0.40 | 0.60 |

upgrading is represented graphically in Fig. 4. Overall, the distributed configuration evaluated delivers 60kt/y of HTL biocrude, and 50 kt/y of hydrotreated biocrude.

*Modelling results*

The results of the modelling section are discussed in terms of the overall mass and energy flows from the different process streams, their elemental composition and their potential utilization/integration within the process. The streams summary from the Aspen Plus ® model can be consulted in the Supplementary material.

Fig. 5 and Fig. 6 show the overall mass and energy balance of the HTL and hydrotreating plants based on the specified product yields. Overall, the configuration results in the production of 50 kt/y of hydrotreated biocrude predominantly in the diesel range (Fig. 7), which from the demand side is very limited and evidences that large-scale production is mainly constrained by the feedstock availability (a medium-size containership (8000 TEU) at average speed (21 knots) consumes about 150 t/d of bunker fuel [20]). In the HTL process, the largest mass flow going through is the aqueous phase, however, the biocrude is the product with the highest energy content. From a biomass input of 100 MW it is estimated that 61 MW are obtained in the HTL biocrude, increasing to 63 MW after the upgrading process. The main by-product in terms of mass and energy flows is the aqueous phase, followed by the solids (mineral product) and the gas product. The differences in mass and energy flows are discussed in the following paragraphs based on the

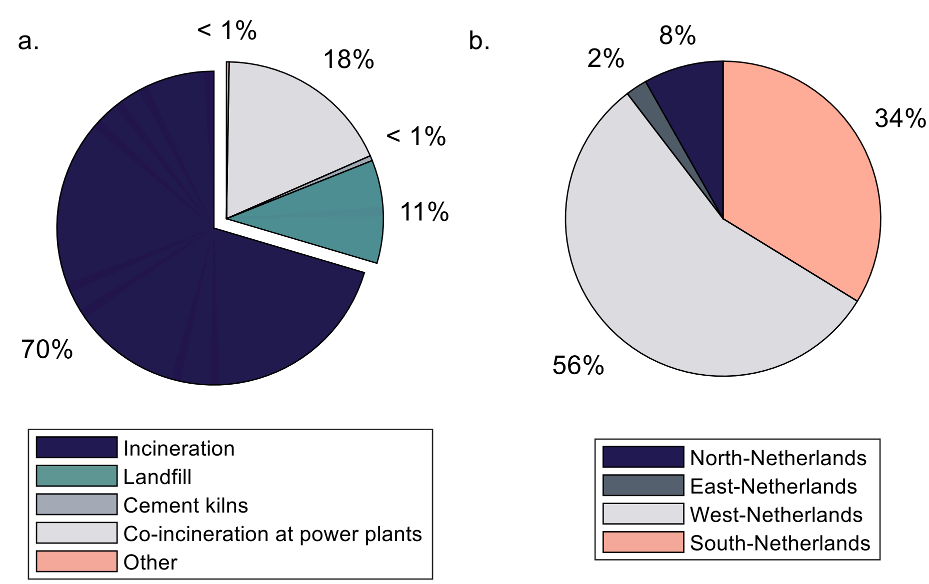



**Fig. 3.** a) Distribution of total sewage sludge produced by disposal method and, b) Regional distribution of sludge incinerated in The Netherlands in 2018 [22].

**Table 6**
Potential HTL biocrude and hydrotreated biocrude production from sewage sludge currently disposed via incineration. Data obtained from [22].

| Region | Wet sludge incinerated in 2018 [kt] | Average dry matter [wt. %] | Average ash (dry basis) [wt. %] | Dry ash free sludge [kt/y] | Dry sludge available per day [t/d] | HTL plants per region | Potential HTL biocrude production [kt/y] | Potential hydrotreated biocrude production [kt/y] |
|---|---|---|---|---|---|---|---|---|
| **Netherlands** | 880.37 | **24.29** | **29.35** | **153.21** | **647** | **6** | **61.28** | **51.48** |
| West | 491.59 | 24.50 | 27.39 | 87.43 | 361 | 3 | 34.97 | |
| South | 297.29 | 24.80 | 30.47 | 51.26 | 221 | 2 | 20.50 | |
| North | 72.05 | 23.31 | 32.70 | 11.30 | 50 | 1 | 4.52 | |
| East | 19.45 | 23.77 | 30.41 | 3.22 | 14 | 0 | 1.29 | |

**Table 7**
Tentative locations of HTL regional plants and host WWTPs.

| | Region | Tentative location HTL plant | Main WWTP capacity [p.e] [23] | Estimated WWTP capacity* [ML/d] | Sludge transport [km]** | HTL biocrude transport [km]*** |
|---|---|---|---|---|---|---|
| 1 | North | Groningen | 213,300 | 42.66 | 50 | 280 |
| 2 | West | Amsterdam | 733,320 | 146.66 | 20 | 80 |
| 3 | West | Port of Rotterdam | 765,000 | 153.00 | 30 | 40 |
| 4 | West | Port of Rotterdam | 423,000 | 84.60 | 15 | 30 |
| 5 | South | Breda | 360,000 | 72.00 | 30 | 50 |
| 6 | South | Eindhoven | 443,700 | 88.74 | 40 | 110 |

*1p.e = 200 L/d of wastewater; ** Average distance within region to main WWTP; *** Distance to Port of Rotterdam.


**Fig. 4.** Simplified representation of distributed configuration of HTL plants and centralized upgrading.

distribution of the different elements (C, H, O, N) across the process, and a more detailed discussion on the energy balance and the different possibilities to harness the energy flows within the process are discussed subsequently.

*Elemental balance and carbon and nitrogen distribution.* The results of the elemental balance across the HTL reactor and hydrotreater are presented in Table 8 together with the elemental composition of the modelled biocrudes. In general, the errors in the elemental balance across reactors are below 1 %, with higher errors in the estimated hydrogen, oxygen and nitrogen contents of the HTL biocrude between 6 and 15 %. Overall, the errors can be explained by the limitations in the model compounds approach in the HTL biocrude and the limited knowledge on the composition of the aqueous phase in both HTL and upgrading reactions. Nevertheless, the errors in higher heating values (HHV) and standard enthalpies of formation ($\Delta h_f^\circ$) are below 3 %, and the true boiling point curves of the modelled biocrudes are in relatively good agreement with the experimental data (Fig. 7), showing satisfactory results that give confidence in the energy calculations. Since the composition of the biocrude model compounds is adjusted following a multi-objective optimization procedure with focus on accurate enthalpy estimation, the observed errors in the distillation profile are expected but still minimized to match other properties simultaneously. The model compounds utilized can be found in the stream summary in the Supplementary material.

The results of the carbon and nitrogen distribution among the different streams in Table 9 show that about 60 % of the initial carbon is recovered in the HTL biocrude and an estimated 56 % ends in the hydrotreated biocrude. The main carbon losses are in the HTL aqueous and solid products that contain approximately 30 % of the initial carbon, while only 8–9 % is lost in the gaseous products of the HTL and upgrading processes, explaining the differences in the energy flows. In terms of nitrogen, most of it is separated in the aqueous phase from the HTL reactor and hydrotreater, with 68 % and 20 % of the initial nitrogen respectively; however, still 0.9 % of the initial nitrogen is obtained as an impurity in the hydrotreated biocrude based on the experimental data used, although recent in-house, yet unpublished experiments have shown that nitrogen can be completely removed by hydrotreatment. These results are in agreement with the typical values reported in literature [4,6].

- Energy integration and by-products utilization

The results of the energy balance show that the highest energy losses are in the aqueous by-product, which highlights the importance of implementing a suitable strategy to harness its potential. The aqueous phase from the process is approximately 100,000 $m^3$/y or 300 $m^3$/d and corresponds to 0.1–0.5 % of the influent, being highly diluted upon recirculation. Under this assumption, the aqueous phase would return to the plant and undergo primary and secondary treatment already in place, consisting in physical separation processes and biological treatment.

Nevertheless, this implementation is subject to the allowable concentrations and parameters of WWTP, so a more detailed evaluation with emphasis in specific entry points in existing wastewater treatment operations must be considered in future evaluations. Based on the carbon and nitrogen distributions the estimated TOC and TN are 18.8 and 13.4 g/L respectively, in line with the TOC between 16 and 27 g/L and TN between 5 and 10 g/L reported in HTL literature of sewage sludge [4,49], but significantly higher than typical carbon and nitrogen concentration in wastewater in the order of 50–500 mg/L. Even though the high dilution factor could be effective in balancing the high loads from the HTL aqueous phase, a key aspect to consider is the type of components present, their bio-degradability and threshold concentrations. Further validation including more detailed data in this regard will be needed to support this configuration, taking into consideration that the

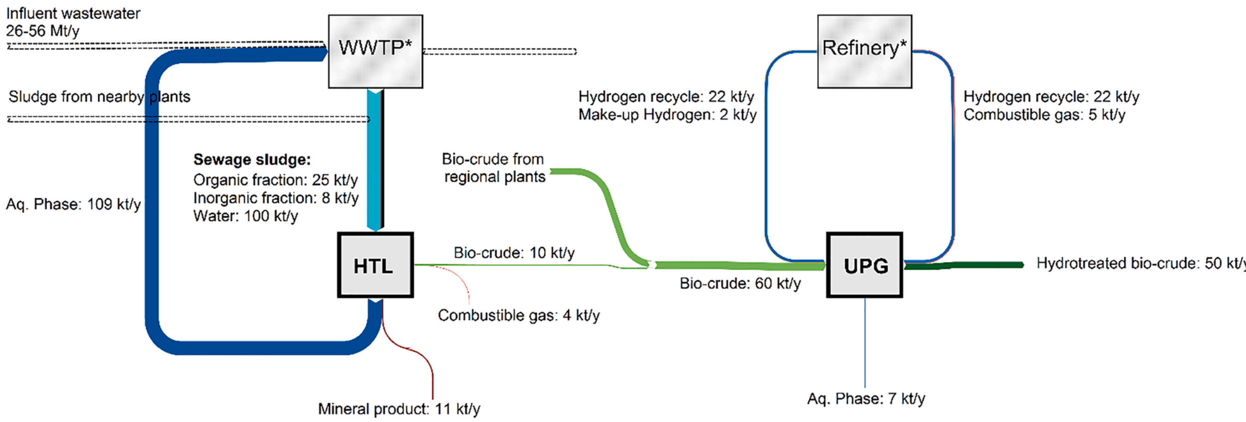



**Fig. 5.** Sankey diagram of mass flows in configuration of distributed HTL plants and centralized upgrading. (*WWTP and Refinery are shown as reference to the co-location scheme but are excluded from the analysis).

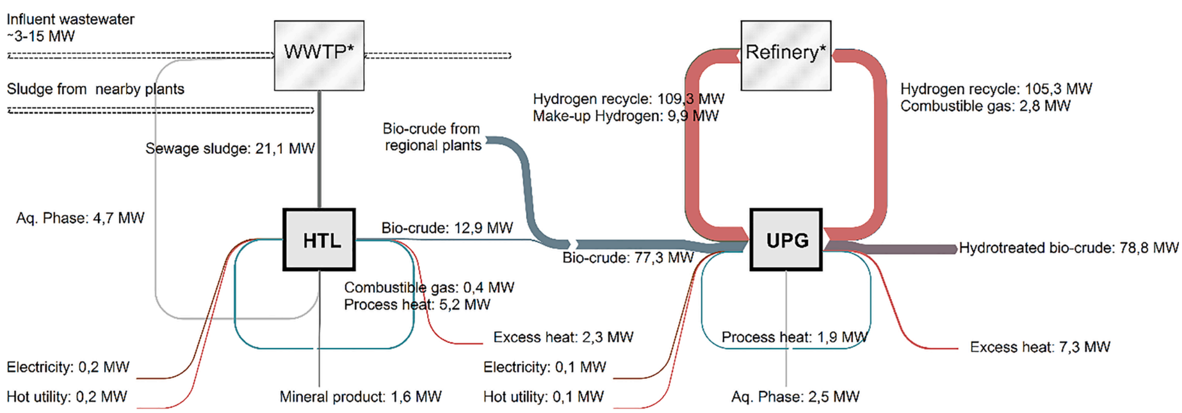



**Fig. 6.** Sankey diagram of energy flows in configuration of distributed HTL plants and centralized upgrading. (*WWTP and Refinery are shown as reference to the co-location scheme but are excluded from the analysis).

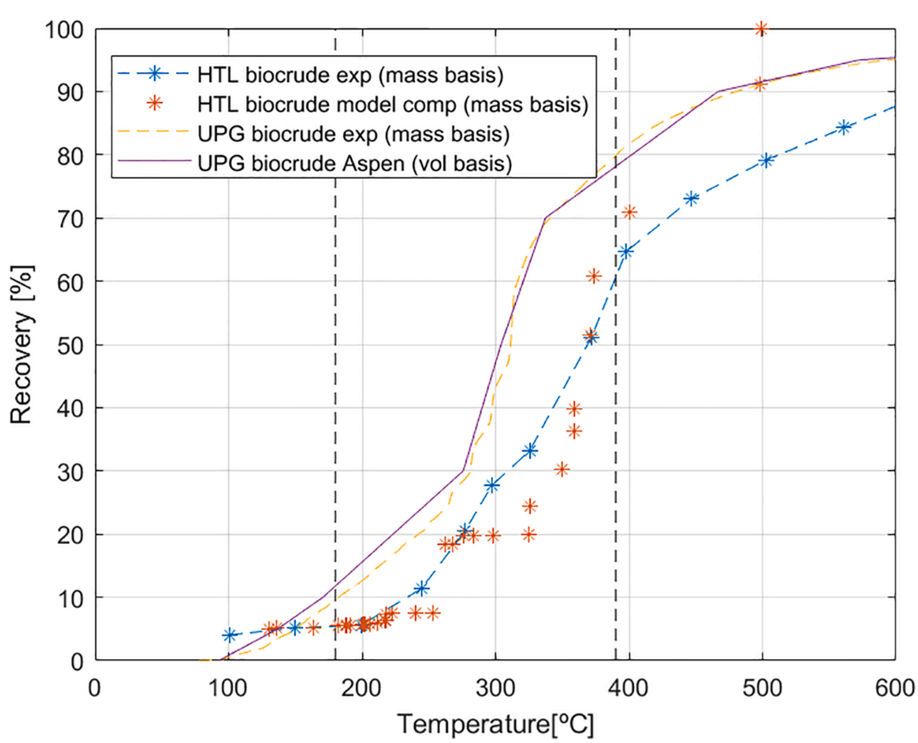



**Fig. 7.** True boiling point curves of experimental and modelled HTL and hydrotreated (UPG) biocrudes.

**Table 8**
Modeling results of the elemental composition of HTL and hydrotreated biocrudes and elemental balance across reactors.

| | Modeling result | | Error [%] | | | |
|---|---|---|---|---|---|---|
| | HTL biocrude | Hydrotreated biocrude | HTL biocrude | Hydrotreated biocrude | HTL reactor | Hydrotreater |
| C [wt. %] | 78.04 | 86.10 | 0.86 | 0.94 | 0.36 | 0.19 |
| H [wt. %] | 10.79 | 13.89 | 16.23 | 0.65 | 1.24 | 0.29 |
| O [wt. %] | 6.05 | 0.00 | 12.79 | 0.00 | 0.02 | 0.00 |
| N [wt. %] | 5.12 | 0.00* | 5.93 | – | 0.38 | 0.00 |
| HHV [MJ/kg] | 38.17 | 45.85 | 1.52 | 0.54 | – | – |
| Δhf° [MJ/kg] | −1.63 | −1.93 | 0.13 | 3.21 | – | – |

*Not included in pseudo-components breakdown.

**Table 9**
Modeling results of carbon and nitrogen distribution among HTL and hydrotreating products.

| | HTL | | Hydrotreating | |
|---|---|---|---|---|
| | C balance [%] | N balance [%] | C balance [%] | N balance [%] |
| Biocrude | 61.24 | 23.99 | 91.93 | 14.78 |
| Gas phase | 8.96 | 0.62 | 7.88 | 0.00 |
| Mineral product | 13.44 | 7.77 | 0.00 | 0.00 |
| Aqueous phase | 16.00 | 68.00 | 0.00 | 85.22 |
| Total | 99.64 | 100.38 | 99.81 | 100.00 |

organic load returned from the HTL to the host WWTP plant increases with the number of WWTPs in the cluster. Other alternatives that have been investigated in literature for the aqueous phase treatment include catalytic hydrothermal gasification (CHG) and distillation for the recycling of the concentrated organics to the HTL reactor [17,50].

Following in energy content, the mineral product from the HTL unit is obtained in significant amounts with about 70 % of the mass corresponding to the original inorganic fraction in the sludge that is not converted through HTL. Besides its carbon and nitrogen content, experimental data reported in literature indicate that the HTL solids concentrate about 95 % of the phosphorous from the sludge [6], having potential to be used in the agriculture sector as fertilizer. Based on the 2018 statistics in [22], the total P in the sewage sludge disposed via incineration was 6588 tonnes, from which the P content in the sludge is estimated to 0.74 wt% on wet basis (or 3.3 wt% dry basis). The total P content in the mineral product is then estimated in 8.6 wt% (dry basis), assuming a 95 % P recovery. Nevertheless, the spread of the HTL mineral product in the fields without undergoing additional treatment is not allowed by current EU legislation, and therefore its disposal is seen in this study as an expense rather than a revenue, either for final disposal or for phosphorous extraction. It is worth to mention that similar products obtained through thermochemical conversion such as pyrolysis and gasification biochars have been recently included in the revised EU fertilizers regulation [51], indicating that there is an opportunity for the HTL mineral product likewise. Alternatively, the mineral product can be combusted to recover any residual heating value, and the resulting ashes treated as fly ash for the extraction of phosphorous, rare-earth metals, etc.

The HTL gas product is mainly composed by $CO_2$ but yet presents a heating value due the presence of combustibles (hydrogen, hydrocarbons, etc.,) that could be utilized within the process. This could be feasible in WWTPs with anaerobic digesters that typically have gas engines installed to burn the biogas produced for heating/electricity purposes, based on the assumption that the ratio of HTL gas to biogas is low and efficiency losses are not significant. If heat integration is implemented and the heating potential of the gas used, it is estimated that only 1 MW of external heating source is needed per 100 MW of biomass input, increasing to 3 MW if the HTL gas is not accounted and to 27 MW without any heat integration. Other alternatives for the produced $CO_2$ could be considered in a more centralized configuration. For example, the $CO_2$ can be captured in-place and transported by truck, taking advantage of its high pressure and concentration in the gas (90–95 vol%), or can be utilized in other industrial applications. The $CO_2$ produced at the mono-incineration plant in Moerdijk is currently delivered to an adjacent plant for the production of calcium carbonate used in paper production [52].

About 10 MW of residual heat is obtained per 100 MW sludge input in a temperature range between 30 and 100 °C, which constitute an energy loss in the absence of other systems such as district heating networks that can make use of some of this potential. Natural gas is the main source of heat in The Netherlands and district heating schemes have only a small share, nevertheless, larger shares are expected in the coming years supporting the transition to renewables. The results also show a relatively low electricity requirement that in the model corresponds only to the energy required for pumping the sludge to the HTL pressure. Compared to the lignocellulosic case, sewage sludge does not require electricity for grinding, but a higher electricity consumption is expected in the downstream processing for emulsion breakup and biocrude separation that is currently not represented in the model.

Regarding the energy balance in the hydrotreater, electricity and external heat supply are relatively low and the main energy by-product leaving the process is the excess process heat. Heat integration in the hydrotreater decreases the estimated external utility from 2.6 to 0.13 MW per 100 MW of biocrude input, while excess heat is produced in a temperature range between 30 and 270 °C. The energy losses in the aqueous phase are significantly lower than in the HTL process and overall reflect the high carbon yield towards the biocrude. From the

**Table 10**
Selected physical properties of hydrotreated biocrude and ISO marine fuels specifications.

| | Hydrotreated biocrude | RMG 180 (380) [20] | | DMA(DMZ) [20] | | DMB [20] | |
|---|---|---|---|---|---|---|---|
| | – | Min | Max | Min | Max | Min | Max |
| Viscosity @ 40 °C [cSt] | 3.36 | – | 180 (380) | 2 (3) | 6 | 2 | 11 |
| Density @ 15 °C [kg/m3] | 833* | – | 991 | – | 890 | – | 900 |
| Flash point [°C] | 62.93 | 60 | – | 60 | – | 60 | – |
| Sulfur [wt. %] | 81.09 ppm* | – | 3.5 | – | 1.5 | – | 2.0 |

*In-house experimental results.

model results, a good carbon closure is obtained even without the inclusion of water-soluble organics with an error below the 1 % (Table 8); nevertheless, remaining carbon and nitrogen, expected in lower quantities than in the HTL case, can be removed before discharge with conventional water treatment at the refinery.

*Drop-in potential of hydrotreated biocrude*

Table 10 presents the results of the physical properties of the HTL biocrude after hydrotreatment in comparison with the minimum and maximum allowed values for residual and distillate marine fuels. The results of viscosity, density, flash point and sulfur content are in compliance with the requirements of both residual and distillate fuels, and based on this, further purification/distillation was deemed obsolete. Regarding the HTL biocrude before hydrogenation, its incompatibility as drop-in fuel has been well documented in existing literature due to its oxygen and nitrogen content, low fuel stability and miscibility, and relatively high viscosity, requiring alternative strategies to be used as fuel blendstocks [53–55]. Still, a detailed validation is necessary in future studies including the evaluation of scenarios with less upgrading requirement, particularly for residual fuels, from a cost-optimization perspective.

*Cost estimation*

The different cost contributors for the production of the HTL biocrude (HTL) and hydrotreated biocrude (HTL + UPG) are shown in Fig. 8 for the base case scenario, with more detailed results presented in Table 11.

For only the HTL process, the production cost of the HTL biocrude is estimated to 1561 EUR/t (42.1 EUR/GJ) but the MFSP is only 230 EUR/t (6.2 EUR/GJ) due to the negative price introduced by the sludge gate fee based on the reference price of mono-incineration. The additional cost of hydrotreating cost is estimated in 380 EUR/t, resulting in an overall MFSP of the hydrotreated biocrude of only 652 EUR/t (14.5 EUR/GJ). In this scenario, the sludge-tipping fee could largely compensate the biocrude production cost.

In the HTL plants, the main cost contributors are the CAPEX and the fixed operational costs (mainly due to high maintenance costs), with 35 % and 32 % of the total respectively, as a result of the low economy of scale associated with relatively small size plants. The high maintenance costs prescribed in the SCENT method (6 % of the fixed capital investment) are in line with other typical factors used in chemical engineering for fluid–solid handling processes [46]. On the contrary, sludge transportation has a relatively small share in the overall costs of only around

**Table 11**
Production costs and MFSP of HTL and hydrotreated biocrude in the base case for sewage sludge case study.

| | HTL | | HTL + UPG | |
|---|---|---|---|---|
| | MEUR/y | EUR/t* | MEUR/y | EUR/t** |
| Annualized CAPEX | 5.5 | 552.9 | 5.0 | 99.5 |
| Hydrogen | – | – | 6.7 | 133.2 |
| Final solids disposal | 1.4 | 143.5 | – | – |
| Aqueous phase disposal | 0.5 | 46.3 | 0.0 | 0.6 |
| Transport cost | 0.9 | 94.0 | 0.9 | 18.2 |
| Utilities and chemicals | 1.1 | 106.6 | 0.7 | 14.5 |
| Fixed operational costs | 5.0 | 500.5 | 4.6 | 91.5 |
| Tax | 1.2 | 117.6 | 1.1 | 21.2 |
| **Total** | **15.6** | **1.561.4** | **19.1** | **378.7** |
| Feedstock | – 13.3 | – 1.331.6 | 13.8 | 273.3 |
| **Total** | **2.3** | **229.8** | **112.8** | **652.0** |

*HTL biocrude basis, **Hydrotreated biocrude basis.

6 %, explained by the relatively short distances to cover within regions in The Netherlands, suggesting that a more favorable balance between plant size and transportation costs can be achieved by increasing the plants size, resulting in higher economy of scale and lower MFSP. These results show that, at current incineration fees, the HTL technology has potential to be a more economical method for sludge disposal with the additional advantage of delivering biocrude for use in transport.

The results support the configuration of several WWTPs feeding the HTL plant and indicate that a plant size of 100 dry t/d of sludge input is close to the minimum capacity to breakeven the sludge disposal costs included without revenues from the HTL biocrude. From this it can be inferred that in The Netherlands the size of individual plants is not enough to implement HTL in an economic feasible way; nevertheless, an evaluation with specific data per WWTP would be needed to assess economic feasibility individually. In a recent study, Seiple et al. estimated that the implementation of the HTL technology in the U.S fleet of WWTPs is economically feasible in facilities with capacities ≥ 17 ML/d based on a sludge/solids disposal cost of 400 USD per wet tonne [18]. In the Dutch scenario a higher minimum capacity can be reasonably expected due to the significantly lower costs reported for sludge and solid residue disposal.

Regarding the hydrotreated biocrude, the results in Fig. 8 show that the final MFSP depends mainly on the price of the supplied HTL biocrude followed by the $H_2$ cost and the CAPEX. In Fig. 8 the dotted line represents the total HTL biocrude production cost (without gate fee), but the MFSP estimation includes the sludge tipping fee, decreasing the final price from 2200 EUR/t (49.7 EUR/GJ) to 652 EUR/t (14.5 EUR/GJ) and

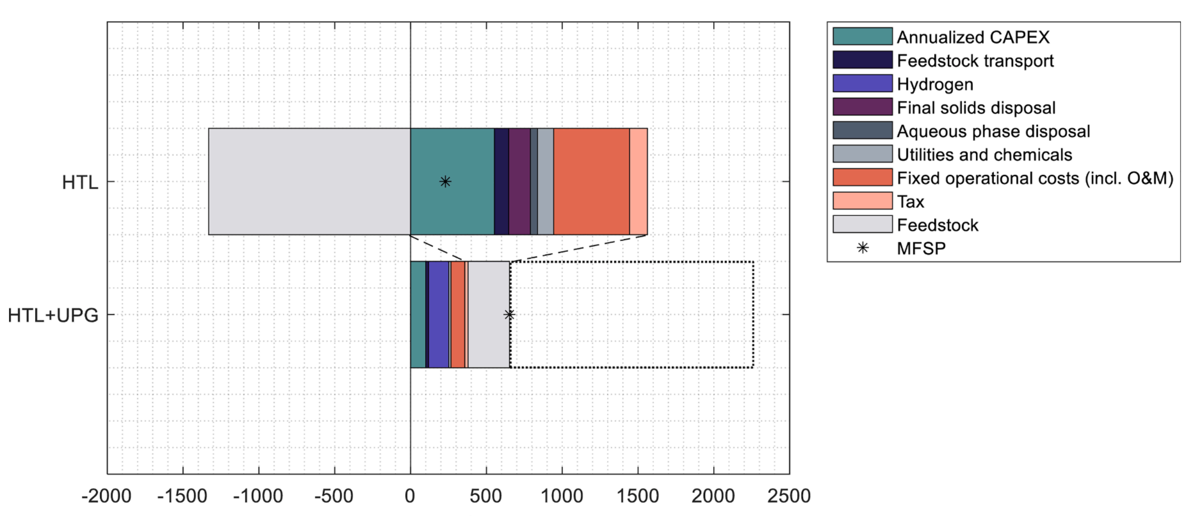



**Fig. 8.** MFSP breakdown of HTL and hydrotreated biocrude in base case scenario.

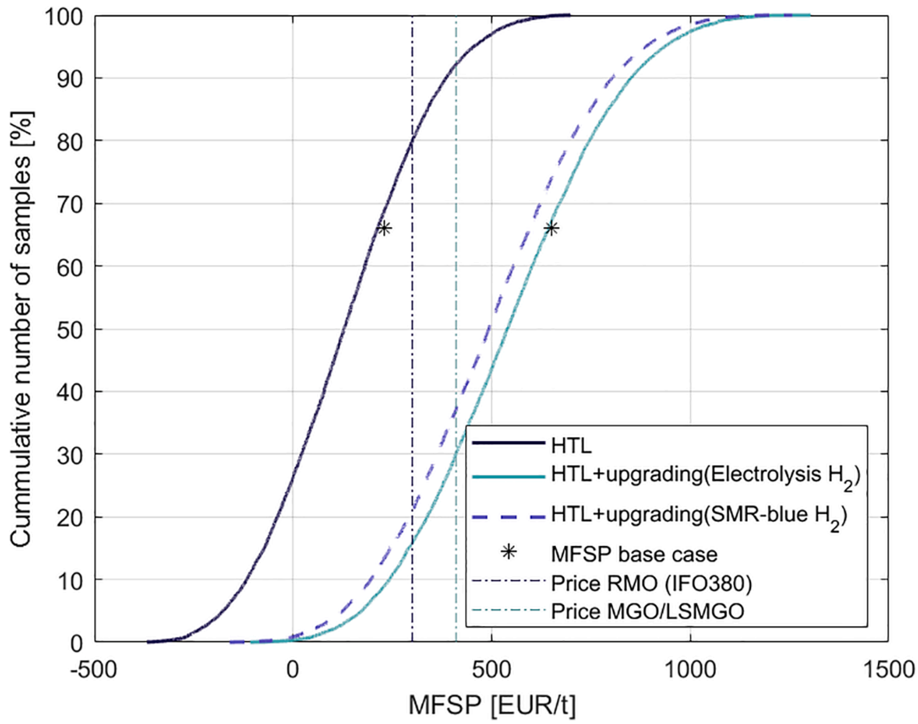



**Fig. 9.** Results of Monte Carlo evaluation of MFSP for HTL and HTL + upgrading.

evidencing the major impact of the sludge credit in the cost analysis.

Finally, the impact of varying the costs parameters in the MFSP is presented in Fig. 9. For some scenarios, the HTL biocrude has a negative MFSP which reflects lower production costs relative to the disposal fee assumed. In case the biocrude is supplied at zero cost, the MFSP of the hydrotreated biocrude can be as low as 400 EUR/t. For the hydrotreated biocrude, the $H_2$ price range was evaluated for two cases ($H_2$ from electrolysis and from steam methane reforming with CCS (blue $H_2$)) using values expected for a Dutch $H_2$ market in [43], indicating that the MFSP can be about 50 EUR/t more expensive in average if $H_2$ from electrolysis is used. Overall, the MFSP estimates are in a competitive price range relative to the current price of marine fuels reported for the Port of Rotterdam around 300 EUR/t of residual fuel (IFO380) and 410 EUR/t of distillate fuel (MGO/ LSMGO) [56].

### *Life-cycle GHG emissions assessment*

The overall results of the GHG emissions assessment are shown in Fig. 10 for the BAU case and the HTL + hydrotreating configuration under the scenarios described in Table 4. Detailed results from

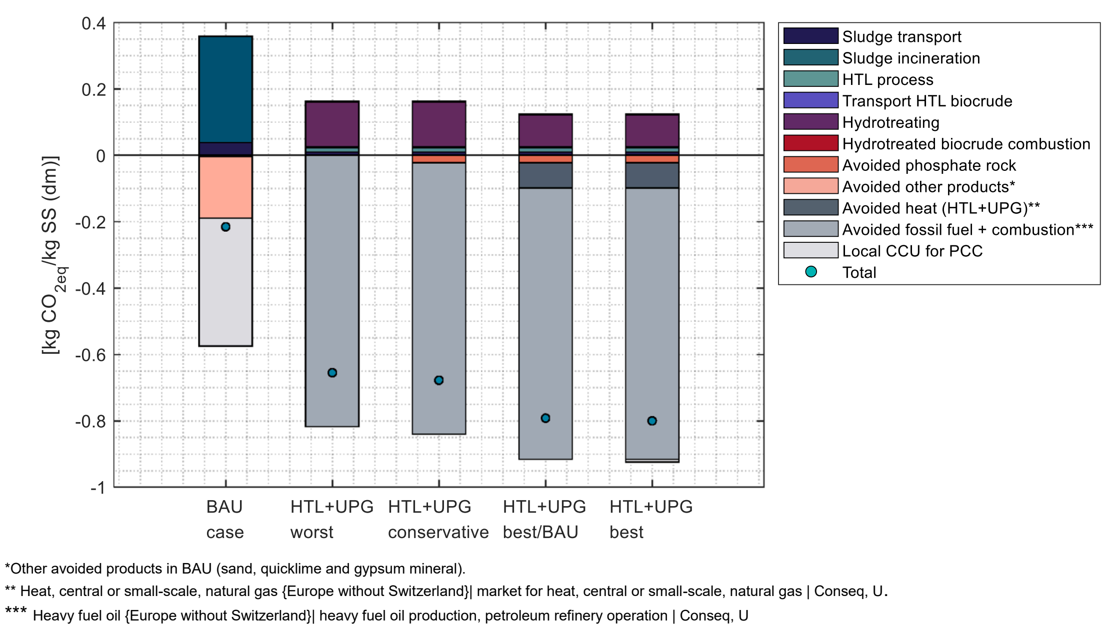



**Fig. 10.** GHG emissions of BAU and HTL + UPG scenarios per unit mass of sewage sludge (dry basis) based on consequential LCA assessment. *Other avoided products in BAU (sand, quicklime and gypsum mineral). ** Heat, central or small-scale, natural gas {Europe without Switzerland}| market for heat, central or small-scale, natural gas | Conseq, U. *** Heavy fuel oil {Europe without Switzerland}| heavy fuel oil production, petroleum refinery operation | Conseq, U.

modelling can be consulted in the Supplementary material. The results show that for the BAU scenario the estimated emissions are −220 g $CO_2$e/kg sludge (dry basis). The modelling is based on inventory data documented by CE Delft [34], which has been re-modelled using the same principles for system expansion and boundary conditions as used in the assessment of the HTL sludge system. This estimate includes the emissions from the sludge incineration, as well as avoided emissions related to utilization of exhaust gas $CO_2$ in a neighboring precipitated calcium carbonate (PCC) plant, and avoided emissions from other by products from the mono-combustion as documented in [34].

For the HTL + hydrotreating scenarios, including the combustion of the marine biofuel product (hydrotreated biocrude) and the avoided production and combustion of marine residual (the substituted product), the estimated emissions are between −650 and −800 kg $CO_2$e/t sludge (dry basis), depending on how optimistically the scenario is modelled. The results indicate a substantial reduction potential in all cases compared to the BAU scenario, mainly due to the avoided emissions from the heavy fossil fuel production and combustion. These have the highest positive impact and largely compensate the emissions associated with the biofuel production process. The results show that the avoided emissions from excess process heat utilization and the avoided phosphate rock are about 10 times lower but seem to be significant in relation to the emissions from the process itself. These are presented in more detail in Fig. 11 and Fig. 12 for the HTL and hydrotreating processes respectively.

For the HTL process (Fig. 11), the results show that the estimated emissions from utilities (heating and electricity) are in the order of 300 kg $CO_2$e/t sludge (dry basis) and can be compensated by the avoided products in the best-case scenarios. The negative parts in Fig. 11 is due to 1) avoided rock phosphate where the processing and transport are the main contributors, 2) avoided heat district heating based on an assumption of reutilization of 60 % of the excess process heat, and 3) local CCU of carbon dioxide from the processing gas exhaust, using the assumptions made for CCU in the BAU scenario [34,35]. The avoided rock phosphate does only reflect the fuels used to extract and transport the mineral, whereas the GHG impact category does not reflect the importance of the avoided resource extraction, which in general are to be the most important issue related to phosphate. The avoided heat (30 % utilization is assumed at a temperature range up to 150 °C) is a reasonable assumption for the future scenario, but the avoided heat will probably be of a lower carbon emission type than the current being dominated by natural gas, which mean that the reality will be closer to the conservative scenario. The local CCU, reflect at current practice at the site for mono-combustion of sludge, but the scalability of this utilization is questionable, which also point towards the conservative scenario.

The hydrotreating process (Fig. 12) has the highest share in the overall emissions due to the hydrogen production via electrolysis and the energy associated with the excess $H_2$ recovery, in total estimated between 950 and 1350 kg $CO_2$e/t sludge (dry basis).The avoided heat in Fig. 12 is based on the possibility of reusing a similar proportion of the waste energy, 60%, avoiding heat-production at the refinery site. As above, this is considered a theoretical and positive potential for emission reductions, but also a reduction that are uncertain, which imply that the conservative HTL-UPG scenario is the most likely one. Still, the results indicate that the HTL + hydrotreating configuration studied has potential to be at least three times more beneficial compared to the BAU scenario from a GHG emissions perspective.

## Conclusions

This study shows promising results from a techno-economic and environmental perspective for the implementation of HTL in a regional network of wastewater treatment plants with centralized biocrude hydrotreating. The results point towards HTL as a potentially more economic technology for sludge disposal compared to mono-incineration in The Netherlands. From the regional configuration evaluated, it can be inferred that in general the size of individual plants in the country is not enough to implement HTL in an economic feasible way, while a more centralized approach is recommended in future studies to further decrease the costs, optimizing the size and location of the HTL plants to decrease capital costs using spatially explicit data.

The parameter with the highest influence in the economic evaluation is the sludge gate fee, and thus the analysis is sensitive to uncertainties in

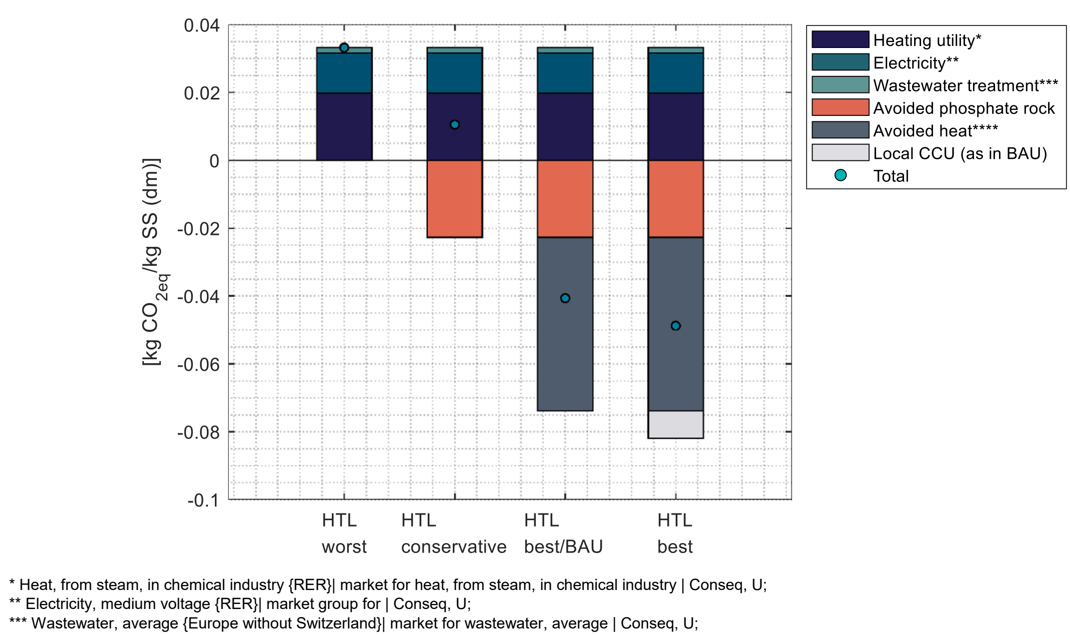
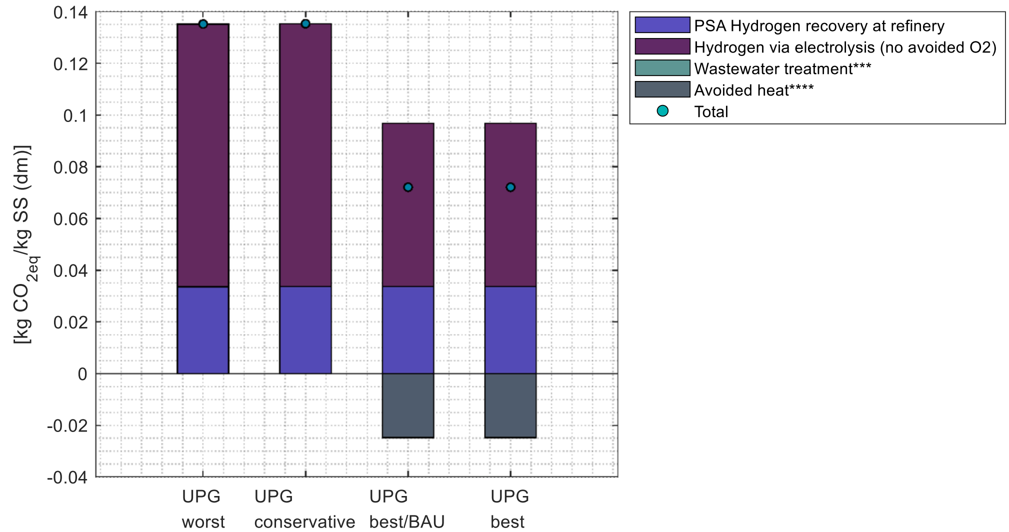



**Fig. 11.** Specific GHG emissions in HTL process based on consequential LCA assessment * Heat, from steam, in chemical industry {RER}| market for heat, from steam, in chemical industry | Conseq, U; ** Electricity, medium voltage {RER}| market group for | Conseq, U; *** Wastewater, average {Europe without Switzerland}| market for wastewater, average | Conseq, U; **** Heat, central or small-scale, natural gas {Europe without Switzerland}| market for heat, central or small-scale, natural gas | Conseq, U.




*** Wastewater, average {Europe without Switzerland}| market for wastewater, average | Conseq, U;
**** Heat, central or small-scale, natural gas {Europe without Switzerland}| market for heat, central or small-scale, natural gas | Conseq, U

**Fig. 12.** Specific GHG emissions in hydrotreating (UPG) process based on consequential LCA assessment *** Wastewater, average {Europe without Switzerland}| market for wastewater, average | Conseq, U; **** Heat, central or small-scale, natural gas {Europe without Switzerland}| market for heat, central or small-scale, natural gas | Conseq, U.

the sludge market and the business/ownership model implemented in the regional scheme and with the refinery. Still, the results show that the business case is likely to be positive for the WWTPs if the HTL biocrude is marketed as a product. Regulatory aspects need to be further considered in this regard as they differ across countries and can represent barriers for technology implementation. Furthermore, in a future scenario of sludge valorization (where it is not seen any more as waste but as resource) its price is likely to switch from negative to positive affecting the business case. Thus, in the long term, more stringent environmental regulations on sludge treatment rather than economic incentives are seen as the main driver for new technologies development, not only in the Dutch case but globally.

Regarding the environmental evaluation, the results indicate that the HTL + hydrotreating configuration studied has potential to be at least 3 times more beneficial compared to the BAU scenario (sludge mono-incineration and fossil marine fuels production) from a GHG emissions perspective. Furthermore, the HTL + hydrotreating configuration holds the potential of a full utilisation of the phosphate content of the sewage sludge. The inclusion of other LCA impact categories such as eutrophication is recommended for future work, given the importance of the nutrients fate in the process.

Despite of being based on data from a specific country, the results of this study indicate the potential of this implementation in other locations with relatively high population density and good transport infrastructure. This can be the case of port areas around the North Sea, projected as green industrial hubs with access to offshore renewable electricity for hydrogen production, where drop-in marine biofuels are expected to play a role with the increasing share of renewables in the marine fuels mix. Particularly on the fuel's characteristics, the properties of the HTL hydrotreated biocrude are indicative of its drop-in potential as distillate marine fuels (DMA/Z and DMB), but other scenarios with less upgrading requirement, particularly for residual fuels, shall be further compared from a cost-optimization perspective and need more detailed validation. In more centralized scenarios of larger HTL plants, the integration of carbon capture in the process is seen as a promising option for BECCS/U implementation, of particular interest in port areas due to recent announcements of CCS projects such as *Porthos* in The Netherlands and *Northern Lights* in Norway.

*CRediT authorship contribution statement*

**E.M. Lozano:** Conceptualization, Methodology, Software, Validation, Writing – original draft. **S. Løkke:** Conceptualization, Methodology, Software. **L.A. Rosendahl:** Conceptualization, Methodology, Writing – review & editing, Supervision. **T.H. Pedersen:** Writing – review & editing, Supervision, Project administration, Funding acquisition.

## 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 data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecmx.2022.100178.

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