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# Life cycle assessment of amine-based versus ammonia-based post combustion $CO_2$ capture in coal-fired power plants

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ABSTRACT

Considering two sets of subcritical and supercritical 500 MW coal fired power plants with and without $CO_2$ capture unit, a comparative, cradle-to-gate life cycle assessment was performed to evaluate the environmental impact of $CO_2$ capture through amine-based and ammonia-based absorption from the flue gasses. The aqueous solutions of monoethanolamine (MEA) was considered as the amine solvent. The effects of MEA concentration of 20, 30, and 40 wt% and $CO_2$ capture efficiency on the environmental impact of different categories were investigated. The methods of IPCC 2013 GWP 100a, AWARE V1.02, ReCiPe 2016 Midpoint (H) V1.03, and TRACI 2.1 V1.05, were considered for this LCA. In terms of global warming, adding a $CO_2$ capture unit presented significant benefits for the environment. However, for other impact categories such as water footprint, ozone depletion, ionizing radiation, marine eutrophication, smog and fossil fuel depletion the plants with $CO_2$ capture unit showed higher impact compared to the plants without a $CO_2$ capture unit. In terms of carbon and water footprints MEA-based $CO_2$ capture unit presents better performance compare to the ammonia-based.

## 1. Introduction

Fossil fueled power plants are considered a main source of carbon dioxide ($CO_2$) emission to the atmosphere (Tong et al., 2018; Zhai et al., 2015). However, these power plants have other environmental impacts such as emission of sulfur oxides, heavy metals, nitrogen oxides and particulates, and also they use water sources (Zhai et al., 2015; Otero-Rey et al., 2003; Yu et al., 2012; Zaid et al., 2019; Zhai et al., 2018; Li et al., 2017). Through various treatment processes most of the above-mentioned contaminants are controlled to the acceptable range. Nevertheless, for the purpose of reduction in greenhouse gas emissions the $CO_2$ capture unit is the main mitigation option. Amine-based absorptions are the most mature technology for post-combustion $CO_2$ capture (Agbonghae et al., 2014; Plaza et al., 2017; Rochelle, 2009). In the amine-based chemical absorption using monoethanolamine (MEA) is a well-established process in industry for the treatment of acid gasses (e.g., $CO_2$ and $H_2S$). The $CO_2$ capture requires additional energy for its operation, which negatively affects the energy efficiency of the power plant. As a consequence, more fossil fuel needs to be used to achieve plant capacity.

The amine-based $CO_2$ capture unit uses water resources and chemicals (e.g., alkanolamine, caustic soda, additives), and produces chemical waste thereby introducing additional environmental impact. The degradation products of the alkanolamines in $CO_2$-loaded solutions can cause corrosion of the equipment which augments the operation and maintenance costs. These degradation products produce chemical wastes which should be treated in a reclaimer and disposed. Accordingly, sulfur dioxide and nitrogen dioxide in the absorber unit react with the MEA solvent to form heat-stable salts and consequently reduce the absorption capacity of the solvent, leading to solvent loss because they cannot be regenerated (Pehnt and Henkel, 2009).

On the other hand, ammonia-based technology presents an alternative way for $CO_2$ capture. In comparison with conventional alkanolamine process, ammonia-based has some advantages such as low cost, no solvent degradation, low regeneration energy, and high $CO_2$ capture capacity (Zhai et al., 2018; Wang et al., 2017; Li et al., 2014; Li et al., 2015; Zhai et al., 2019; Li et al., 2016). Depending to the temperature in the absorber ammonia-based process can make some kind of precipitates (Darde et al., 2009). The reaction of ammonia with $CO_2$ can produce different precipitation products such as ammonium bicarbonate ($NH_4HCO_3$), ammonium carbonate ($(NH_4)_2CO_3,H_2O$), ammonium carbamate ($NH_2COONH_4$), and sesqui-carbonate ($(NH_4)_2CO_3$,

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$2NH_4HCO_3$) (Darde et al., 2009). Considering ammonia volatility and the precipitation products of $CO_2$—$NH_3$—$H_2O$ system, makes the ammonia-based $CO_2$ capture much more demanding (Yang et al., 2013). Therefore, it is important to understand the big picture by evaluating various scenarios in the $CO_2$ capture unit's overall environmental impact during its life cycle.

In the case of MEA aqueous solutions its 30 wt% (≈MEA 7 m) is usually considered as the conventional and base case for $CO_2$ capture in absorption units (Fisher et al., 2007). However, considering the plant performance, capital and operational cost, flue gas composition, amine concentration effect on the degradation products, amine and its degradation products toxicity and corrosive properties, a range of MEA concentrations mostly between 20 and 40 wt% can be applied for the $CO_2$ absorption (Rochelle, 2009; Aronu et al., 2011; Zhang et al., 2011; Huertas and Garzon, 2015; Dhingra et al., 2017).

Huertas, et al., discussed the amine concentration dependency of the $CO_2$ absorption capacity of MEA solutions. They showed that the $CO_2$ absorption capacity can change between 0.45 to 0.47 kg $CO_2$ /kg MEA, for MEA 30 and 20 wt%, respectively (Huertas and Garzon, 2015). Accordingly the kg $CO_2$ /kg MEA value increases up to 0.58 kg $CO_2$ /kg MEA for MEA 2.5 wt%,which means that high MEA concentration provides lower $CO_2$ absorption capacity per mass of amine in the solution. However, as can be deduced, in the operational range of MEA concentrations from 20 to 40 wt% in $CO_2$ capture units, no significant change in $CO_2$ absorption capacity in kg$CO_2$/kg MEA is expected. However, despite relatively similar capacity of MEA solutions for $CO_2$ absorption per amine mass in the solution in operational MEA concentrations, the concentration variations may significantly affect the environmental impact of the capture process.

Nowadays, Life Cycle Assessment (LCA) is the most inclusive approach to study the environmental impact of products, technologies or processes (Guinée et al., 2011; Tabone et al., 2010; ISO 14040 2006a; ISO 14044 2006b). During last two decades several LCA studies have been performed and reported for various $CO_2$ capture technologies including amine based processes (Pehnt and Henkel, 2009; Lombardi, 2003; Koornneef et al., 2008; Nie et al., 2011; Schreiber et al., 2009; Singh et al., 2011; Zhang et al., 2014; Zhou et al., 2014). A comparative LCA study for $CO_2$ capture technologies for advanced supercritical plant with MEA, and with AMP/PZ and cryogenic oxy-fuel, was performed by Oreggioni et al. (2017). Their results indicate no remarkable variations in climate change potential for the technologies under their study. However, in terms of other environmental issues such as acidification, toxicity, resource depletion, their results showed an increment as a result of the $CO_2$ capture unit.

Grant et al. (2014) performed a comparative LCA for the $CO_2$ capture between the use of MEA and a potassium carbonate based solvent. In a similar study a comparative LCA was done for CSI ($CO_2$ Solutions Inc, technology for $CO_2$ capture based on stable salt solution plus carbonic anhydrase enzyme as a catalyst for carbon management), MEA and UNO MK3 (Saunier et al., 2019). Koornenef et al., used LCA to evaluate the environmental impacts of subcritical and ultra-supercritical pulverized coal fired electricity generation with and without carbon capture and storage (CCS) through a cradle to grave study. They considered MEA solvent for $CO_2$ capture. They showed that, $CO_2$ capture unit can reduce the GHG emissions are lessened significantly to 243 g CO2-eq/kWh (Koornneef et al., 2008).

Clarens et al., implemented a comparative Cradle-to-gate LCA to study the environmental impact of $CO_2$ capture from the flue gasses in a subcritical coal power plant between three technologies. They considered two types of amine processes including MEA and Econamine FG+ (advanced amines) and CaO looping their study. They showed that carbon dioxide emissions were 0.33, 0.27 and 0.26 kg $CO_2$-eq/kWh for, Econamine, Econamine FG+and the CaO looping capture unit, respectively, compared to the same plant without a capture unit (0.96 kg $CO_2$ -eq/kWh). In terms of environmental perspective, they mentioned that with the assumptions and system boundaries defined in their study, the CaO looping can be a potential and a feasible option to amine-based systems (Clarens et al., 2016).

An LCA of $CO_2$ capture and storage (CCS) for lignite power plant technologies was performed by Pehnt and Henkel (2009). Their LCA study included post-combustion, pre-combustion and oxyfuel capture processes. Their study showed a significant decline in greenhouse gas (GHG) emissions but an increase in cumulative energy demand for all $CO_2$ capture approaches in comparison with power plants without CCS. Nevertheless, they mentioned that the feasible CO2 capture technologies for the post-combustion accompany with prompt increases in many impacts' categories, while the only acidification is reduced. They discussed that for the oxyfuel process the result of the LCA depends particularly on two parameters: the energy demand for air separation and the feasibility of co-capture of pollutants other than $CO_2$. They mentioned that if co-capture was viable, oxyfuel could potentially end up to almost zero emission power plant.

In a review paper by von der Assen et al. (2014) some important aspects of LCA of $CO_2$ capture and utilization were discussed, such as system boundaries, data collection and environmental impact calculation, as well as interpretation and sensitivity analysis of the LCA results. They mentioned that an environmental assessment using LCA for $CO_2$ capture and utilization (CCU) is necessary and attainable, even early in the development stage. They discussed that LCA is unable to determine utter environmental sustainability thresholds of products but is able to establish environmental hot spots in a product's life cycle and if it is more environmentally beneficial than another.

Manuilova, et al., performed an LCA study of the of the lignite coal fired electricity generating station with and without post-combustion $CO_2$ capture at the Saskatchewan Power Corporation's (SaskPower) Boundary Dam Power Station in Estevan, Saskatchewan, Canada. Their LCA study presented a cutting in global warming and air impact categories, while, escalating in some categories associated with soil and water. They concluded that the $CO_2$ capture process reduces the atmospheric emissions. On the other hand the waste streams became more concentrated due to $CO_2$ capture process as releases to soil and water (Manuilova et al., 2014).

A comparative LCA was performed comparing membrane separation and chemical absorption processes using MEA for post-combustion $CO_2$ capture (Giordano et al., 2018). As expected the study showed that LCA results of $CO_2$ capture based on a membrane separation process were highly depended to membrane material and thickness of the dense active layer, which in turn affect the net power consumption and membrane area requirement.

Nie, et al., performed a comparative LCA study between oxy-fuel and post-combustion $CO_2$ capture using MEA solvent based on their own custom made model (Nie et al., 2011). They showed that the post-combustion and oxy-fuel combustion CCS are able to reduce the global warming potential (GWP) by 78.8% and 80.0% respectively compared to conventional power plant without CCS. However, their analysis presented that other environmental impacts, such as ecotoxicity, human toxicity and acidification, considerably depends on the $CO_2$ capture procedures utilized.

Korre, et al., in an alternative work performed custom life cycle modeling on post combustion $CO_2$ capture with various solvents such as MEA, Hindered Amine KS-1, and promoted Potassium Carbonate K+/PZ (Korre et al., 2010). They showed that, in comparison with plants without capture unit, the post-combustion $CO_2$ capture can reach to approximately 80% reduction in GWP without a notable increase in other environmental impact categories.

A comparative LCA study between bio-catalyzed and promoted potassium carbonate processes and amine-based carbon capture technologies was implemented by Saunier et al. (2019). They did cradle-to-gate LCA of capture processes including CSI, MEA and UNO MK3 (a precipitating potassium carbonate separation process) for the flue gas stream of a 550MWcoal-fired power station in the Midwestern USA. The Impact 2002+ method was considered for the analysis., which contemplates

climate change, human health, ecosystem quality and resources impacts at endpoint level, and sixteen other indicators at midpoint level such as aquatic acidification and Ozone formation. The LCA outcomes present that the CSI process provides remarkably lesser environmental impacts than the MEA and UNO MK3 units.

Although MEA is a well-known alkanolamine for the $CO_2$ capture process, to the authors' knowledge there has been no investigation in terms of comprehensive environmental impact of using different concentrations of aqueous solutions of MEA solvent in $CO_2$ capture. In this work the LCA study of subcritical and supercritical coal-fired power plants with and without capture unit is investigated. The $CO_2$ capture unit selected for this is studied under three different MEA concentrations: 20, 30 and 40 wt% of MEA in its aqueous solutions. The effect of capture efficiency on the LCA results of the power plant will be investigated as well. The environmental impact of ammonia-based $CO_2$ capture unit under subcritical power plant is finally compared with MEA 30 wt% through corresponding LCA. Ammonia 30 wt% solution is selected for this study (Darde et al., 2009). In this way, the MEA concentration effect on the plant environmental impact and its comparison with ammonia-based CO2 capture through LCA results will be presented.

## 2. Methodology

The LCA methodology is a powerful tool to quantitatively and comprehensively evaluate the environmental impacts of a product, process, or technology. LCA provides vital support for decision making in terms of environmental sustainability. According to ISO 14,040 in order to perform the LCA study, four important steps should be included: (a) Goal and scope definition, (b) inventory analysis, (c) impact assessment and (d) interpretation (ISO 14044 2006b).

### 2.1. Goal and scope of the study

The goal of this LCA is to compare the environmental impact of a subcritical (and supercritical in the case MEA solvent) coal-fired power plant with and without a $CO_2$ capture unit. The main target impact will be $CO_2$ equivalent emission and the water footprint. However, other impact categories will also be evaluated in this LCA study. Two important $CO_2$ capture system parameters including amine (i.e. MEA) concentration and capture efficiency effect on the plant environmental footprints over different categories are discussed. The three different concentrations of MEA will be considered (20, 30 and 40 wt%) to comparatively evaluate the environmental impacts of changing the amine concentration in the $CO_2$ capture unit, while maintaining the capture efficiency at the same level (90%). For MEA 30 wt% solvent the $CO_2$ capture efficiency effect on carbon and water footprints is evaluated as well. At the final step the performance of ammonia-based $CO_2$ capture unit with $NH_3$ 30 wt% compare to MEA 30 wt% in terms of the overall plant carbon and water footprint in a subcritical coal-fired power plant will be evaluated. The fuel input is kept constant at 173 and 163.5 ton/hr for subcritical and supercritical power plant, respectively, therefore, the plants electrical output will be changed, Table S9. Due to the comparative nature of this study and assuming the similarity of the power plants, the facility construction and transportation were excluded from the overall analysis since they can be presumed to be equivalent.

The functional unit, which is the basis for an LCA study ensuring that comparisons are functionally equivalent, is defined as the production of net electrical power of 1 kWh by the power plant. The $CO_2$ capture unit consumes a portion of generated steam in the power plant for solvent regeneration and it also needs some additional auxiliary electricity. Therefore, addition of the $CO_2$ capture unit imposes an energy penalty on the plant capacity such that the net power output of the power station is further reduced from the plant's nominal net electrical output. In this study, we have tried to keep fuel input to the plants as a constant value for all scenarios, i.e., plants with and without a $CO_2$ capture unit. Figs. 1 and 2 present the system boundaries and stages included in this LCA. The system boundary includes supply chain for the raw materials to each unit, the base plant (i.e., boiler and in furnace NOx control), NOx control (Selective Catalytic Reduction-SCR), total suspended particulate control (TSP), flue gas desulphurization (FGD), $CO_2$ capture (in the plants with $CO_2$ capture) and compression. In this LCA model, all emissions to atmosphere from the different process units including SCR, FGD, $CO_2$ capture, etc., are gathered in the stack unit, so only the net emissions from the stack unit are relevevant, and there are no emissions to the atmosphere from any of the individual process units. The emissions from amine reclaimer residue was considered in the waste treatment section. Therefore, chemical waste of reclaimer bottom was taken into account under waste end of life in the LCA model. For each power plant, the full

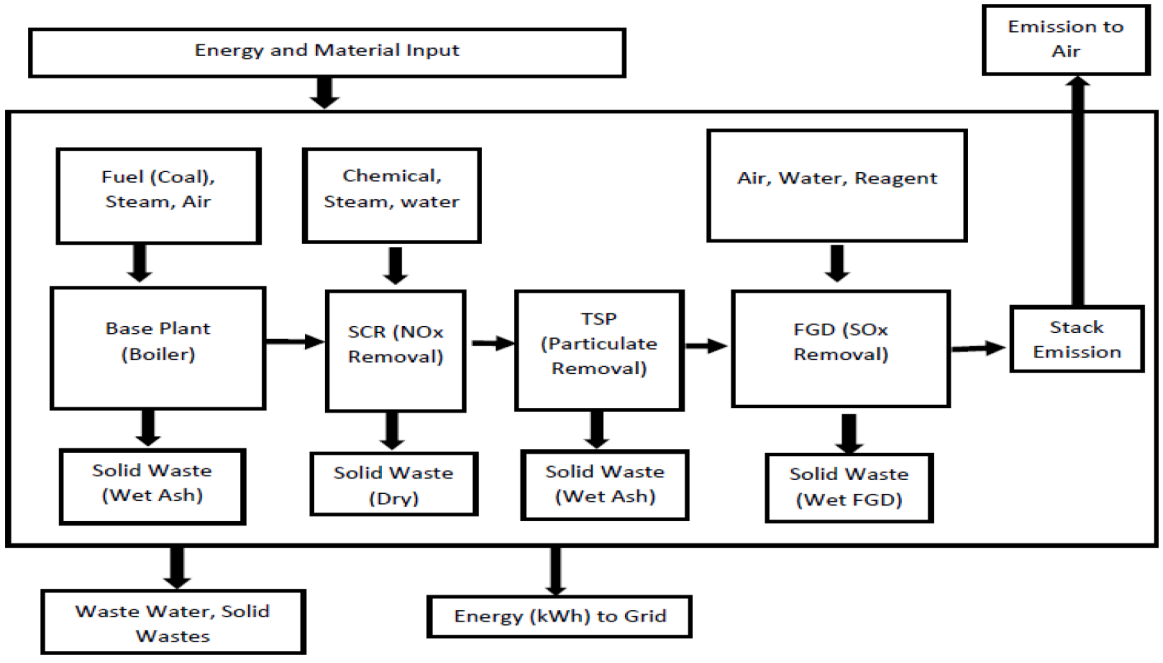


**Fig. 1.** System boundaries, coal fired power plant without $CO_2$ capture.

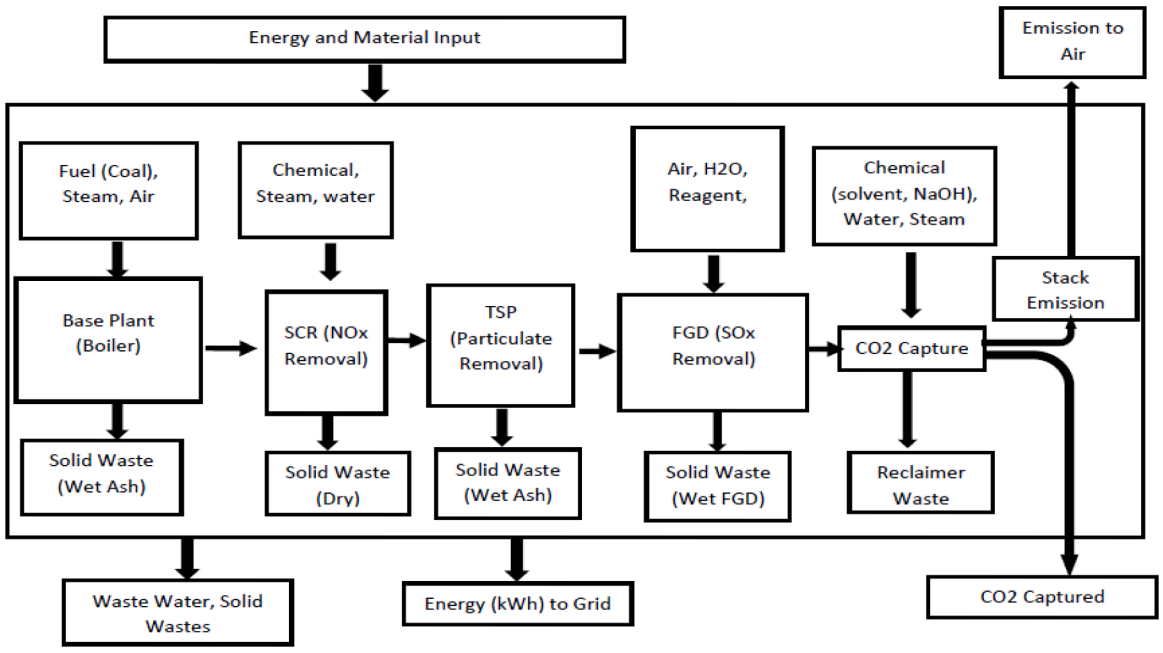


**Fig. 2.** System boundaries, coal fired power plant with $CO_2$ capture.

LCA model includes all supply chain available in the SimaPro – ecoinvent database. Since each unit in the plant modeled separately, then whole related supply chain and energy flow due to auxiliary loads was considered in the unit process of that unit. For instance, to model the boiler (base plant) the process for coal mining/production, the process for auxiliary electricity required for the plant were included in the base plant unit process. Or for the CO2 capture unit, the processes for MEA and NaOH production, the auxiliary electricity used by, all were included in CO2 capture unit process. Therefore, generally, for each unit in the plant, any raw materials or energy used in that unit were considered in the LCA modeling of that specific unit. Eventually, all unit processes were gathered in overall LCA to model the whole plant. In this study, the required electrical inputs (auxiliary loads) of various units were provided from natural gas based sources. Because of the comparative nature of this LCA and the similarity of different units in plants under study, the construction of process units, and transport of the materials, tools, or units were excluded from the analysis. Wastes from FGD consisted of gypsum and limestone. Waste disposal impacts were modeled based on waste treatment processes for gypsum and limestone residue in the ecoinvent 3.5 database (ecoinvent 2020). In this way, the environmental impacts of main wastes including dry and fly ash, FGD waste and the reclaimer waste of $CO_2$ capture unit were included in the model and estimated.

Tables S1 through S7, in supporting information, present life cycle inventory including the data (input/output) generated by IECM software and used in this study (Rubin, 2008). Tables S8–S10, supporting information, present unit processes and overall plants performances, efficiencies and electrical requirements.

### 2.2. Life cycle inventory analysis

The data used in this Life Cycle Assessment (LCA) were extracted from the Integrated Environmental Control Model (IECM) software. This software, developed by the Carnegie Mellon University for the U.S. Department of Energy (USDOE) allows estimating the performance, emissions, and cost of alternative power generation technologies and emissions control options for coal and natural gas power plants. Therefore, data are considered as secondary data source (Rubin, 2008).

For each plant and for the processes located inside that plant (within the system boundaries) separate sets of process units were defined. For the end of life of plant wastes, as is mentioned existing processes in the ecoinvent database for similar main waste materials were chosen. In this way, the environmental impacts of main wastes including dry and fly ash, FGD waste and $CO_2$ unit reclaimer waste were included in the model and estimated through the existing processes.

The material, energy and waste flows for each unit process within the system boundaries were generated by IECM software (Rubin, 2008). It was assumed that the results extracted from the IECM software, in terms of the total plant and each individual unit processes are reliable for such comparison.

The network diagram for selected LCA models from subcritical and supercritical plants using IPCC 2013 GWP 100a are shown in Figs. S1–S4, Supporting Information.

### 2.3. Life cycle impact assessment

The $CO_2$ capture unit presents a net meaningful (>50%) reduction in $CO_2$ emission. Different impact categories include IPCC 2013 GWP 100a (for $kgCO_2$ equivalent) (IPCC 2013), Water footprint (AWARE) (Boulay et al., 2018), ReCipe 2016 Midpoint (H) (Goedkoop et al., 2008; Huijbregts et al., 2016) and TRACI 2.1 (Bare, 2011) methods were considered for this comparative LCA study. The LCA analysis is discussed in the Results and Discussion section.

### 2.4. Interpretation

In this study the secondary data type generated by IECM software were used for the LCA. No evaluation on the data accuracy other than literature citation for the IECM software reliability was performed. However, the LCA results uncertainty and sensitivity over some of major parameters in the plants with $CO_2$ capture were investigated. Nevertheless, for more comprehensive study some selected data need to be verified by experimental data. The LCA shows meaningful impact of $CO_2$ capture unit and amine sorbent concentrations over global warming potential and water use in the processes.

The sensitivity analysis over the percent of gypsum in the waste of FGD unit was performed.

### 2.5. Model and calculation description

The LCA model was performed by SimaPro, 9.0.0.48 release (Sima Pro-LCA Software 2021), using ecoinvent v3.5 database. The material and energy data inventory were provided through IECM software (Rubin, 2008). Nevertheless, for the process waste and their emission to air and water, a combination of available literature, industrial and IECM output was employed. The emission to air from MEA degradation (mainly ammonia) was extracted from IECM model output, while the amine degradation related waste accumulated in the bottom of reclaimer was adapted from relevant experimental, pilot, or plant data available in open literature and reports, which will be discussed in the next section. To perform LCA for each plant the two sets of processes were constructed, including material and waste treatment processes. The various unit processes included in the system boundaries such as base plant, SCR, TSP control, FGD and $CO_2$ capture units were located in the material section of SimaPro software. On the other hand, the waste materials and their landfill were provided in the waste treatment section of software. The whole life cycle for each plant consists of both material and waste treatment sections, Figs. S1–S4, Supporting Information. The gypsum, limestone, wastewater, and average incineration residue were chosen from the ecoinvent database for the FGD waste. For the fly and dry ash control, the fly ash and scrubber sludge treatment process in the ecoinvent database was chosen. In terms of the CO2 capture unit, emissions to air were included in the total stack emission, while for the reclaimer bottom waste separate landfill disposal was considered.

As discussed by Sexton, et al., the metal (e.g. mercury) content of thermal reclaimer waste in coal-fired power plants might result in the wastes being categorized as hazardous wastes (Sexton et al., 2014). Currently in the United States, for the amine reclaimer residue, landfill disposal and/or incineration are the most common methods employed (Sexton et al., 2014). In this study the solid waste disposal through landfill was considered for amine reclaimer sludge. Nonetheless, since $CO_2$ capture unit and solid waste treatment of amine reclaimer sludge is relatively a new process in power plants, sustainable disposal methods might be a requirement (Sexton et al., 2014). As mentioned, IECM provides no information in terms of waste composition in $CO_2$ capture unit. Therefore, in this study, to evaluate the environmental impact of reclaimer residue in landfill disposal, the chemical composition of reclaimer bottom waste were estimated using literature reported values (Veltman et al., 2010; Strazisar et al., 2003; Sexton, 2014). The amine degradation products and heat-stable salts (HSS) are the major contaminants in the amine solution. The oxidative degradation products of MEA can produce organic acids which are the main precursor of HSS. During the $CO_2$ capture process, a slipstream of amine solutions with accumulated degradation products are taken to the thermal reclaimer. Through application of sodium (or potassium) hydroxide solution in thermal reclaimer, a part of HSS can be converted to their precursor anion sodium (or potassium) salts and release the free amine molecules (Veltman et al., 2010; Flø et al., 2017). Formate, sulfate, and nitrate are main HSS products for MEA solution (Thompson et al., 2014; Thitakamol et al., 2007). In this study to be able to take into account major HSS and MEA degradation products in the LCA modeling, in some cases which exact chemical compound (i.e. degradation products) were not available in ecoinvent, the most similar accessible compounds were selected to estimate the environmental impact. The type of degradation products and HSS and their mass fraction in the reclaimer residue were estimated from literature and normalized so that the mass balance of IECM calculated reclaimer waste to be satisfied as close as possible (Veltman et al., 2010; Strazisar et al., 2003; Thompson et al., 2014). Because of low vapor pressure of degradation products in the reclaimer the emission to air is insignificant (Veltman et al., 2010). However, ammonia as one of the major products of MEA degradation with high vapor pressure was considered in this LCA modeling in stack emission to atmosphere. In much (i.e. order of magnitudes) lower extent some volatile aldehydes (e.g. acetaldehyde and formaldehyde) and even in lesser extent nitrosamines can release to atmosphere along with flue gas from absorber (Veltman et al., 2010; Thompson et al., 2018; Thompson et al., 2017). However, it has been mentioned in literature, considering magnitude of aldehydes emissions, they have approximately no impact on various environmental categories (Veltman et al., 2010; Strazisar et al., 2003). In terms of nitrosamines, due to low volatility their emission to atmosphere is expected to be not an issue, especially when primary amines (e.g. MEA) are being used as a solvent in $CO_2$ capture process, and their production rate in MEA as a primary amine are very low (Thompson et al., 2018; Dai et al., 2012; Grebel et al., 2006). On the other hand, taking into account of very low concentration of nitrosamines formation in the liquid phase and their low Henry's constant values, existing water-wash systems are able to successfully capture them prior to entering to atmosphere (Thompson et al., 2018; Thompson et al., 2019). Nevertheless, nowadays there is a concern of environment pollution due to nitrosamine emissions. At the same way, concentration of formaldehyde in MEA solution of $CO_2$ capture unit is usually more than two order of magnitude higher than nitrosamines. Therefore, despite of their extremely low concentrations in the amine solution, for more comprehensive investigation of environmental impacts of $CO_2$ capture units in power plants, both nitrosamine and formaldehyde emission to water through amine reclaimer waste landfill were considered in this LCA study. The quantitative values of formaldehyde and nitrosamines rate of accumulation in MEA solution, were estimated from literature (Dai et al., 2012). The chemical wastes accumulated in MEA solution taken to reclaimer, including degradation products, heavy metals and HSS investigated in this work are listed in Table 1.

**Table 1**
Reclaimer waste composition used in this study.

| Compound [a] | MEA 30% [b] wt% |
|---|---|
| MEA | 35.88 |
| NaOH | 21.36 |
| Formate | 1.24 |
| Sulfate | 19.08 |
| Nitrate | 12.65 |
| Nitrite | 1.04 |
| Chloride | 2.70 |
| HEIA [c] | 2.90 |
| triHEIA [c] | 1.04 |
| HEEDA | 0.41 |
| HEI [c] | 1.66 |
| Hg | 8.92E-04 |
| Se | 1.14E-03 |
| As | 1.87E-04 |
| Cd | 8.30E-05 |
| Cr | 2.24E-03 |
| Pb | 2.28E-04 |
| total - N-nitrosamine [c, d] | 1.99E-04 |
| Formaldehyde [d] | 1.56E-02 |

[a] Chemicals and their amount in the reclaimer waste were mainly adapted from work done by Sexton, 2014. Then data normalized for this study accordingly to be consistent with total output mass of reclaimer waste obtained from IECM. The value, and in most cases the order of magnitudes of data rechecked and reevaluated using other references. Veltman et al., 2010, Thompson et al., 2014, Thompson et al., 2018.
[b] In terms of other concentrations of MEA employed in this study (i.e. MEA 20 and 40 wt%) the wt% of the reclaimer contaminants was calculated only by multiplication of the amine proportional amount in numbers of Table 1.
[C] For these compounds no exact match was found in software database. Therefore, the closest available compounds in ecoinvent database were selected, including, 2-Imidazolidinone for HEIA, 2-Imidazolidinone, 4,5-dihydroxy-1,3-bis(hydroxymethyl) for triHEIA, 1h-imidazole, 2-methyl for HEI, and Nitrosamine, diallyl, for total N—Nitrosamine.
[d] Values estimated from Dai et al., 2012.

## 3. Results and discussion

*Amine-Based* $CO_2$ *Capture:* A conventional amine-based $CO_2$ capture process is comprised of an absorption and desorption cycle in which, using a suitable sorbent, the $CO_2$ is separated from the stream of flue gas and absorbed into the solvent, and then is released from the sorbent as pure $CO_2$. The regenerated sorbent is returned to the absorption column to repeat the cycle. This regeneration step is very energy intensive. The overall reaction between amine (i.e., primary and secondary alkanolamines) and $CO_2$ in an aqueous phase is;

$$2Am + CO_2 \leftrightarrow AmCOO^- + AmH^+$$

Where, Am, $AmCOO^-$, and $AmH^+$ represent amine, amine carbamate and protonated amine, respectively. Using aqueous solutions of alkanolamines (particularly MEA) makes it possible for capturing $CO_2$ at low concentrations (e.g., around 15% in coal power plants).

The IECM software provides the possibility of defining the geographical location of the power plant to be modeled. In this study the US Midwest States including IA, IL, IN, KY, MI, MN, MO, ND, NE, OH, SD, WI and WV were considered for the modeling with the plants' lifetime of 30 years.

Fig. 3 presents the global warming potential (GWP) as kg $CO_2$-eq produced per kWh (kg$CO_2$-eq/kWh) for each plant. As can be seen, in general the supercritical plants provide relatively lower $CO_2$ emissions per kWh compared to corresponding subcritical plants. The addition of a $CO_2$ capture unit further reduced the global warming potential in all cases. According to the results presented in Fig. 3, at the similar carbon capture efficiency (i.e., 90%) the more concentrated amine solvents remove more $CO_2$ per kWh of the plants, and therefore provide better performance in terms of the $CO_2$ capture. The power plant with a capture unit using solvents with 40 wt% MEA produces around 10% less $CO_2$ per kWh energy produced compared to a capture unit with 20 wt% MEA. Table S11, Supporting Information, provides the kg $CO_2$-eq/kWh calculated for each plant. The values of kg $CO_2$-eq per kWh calculated in this work for the sub and supercritical coal-fired power plants without $CO_2$ capture unit, i.e., 0.94 and 0.89, are in close consistency with literature values, i.e. between 0.85 to 1.1 kg $CO_2$-eq/kWh (Clarens et al., 2016; Policy and Analysis, 2016; Spath and M., 2004; Dang et al., 2015; Agrawal et al., 2014). Accordingly, in terms of power plants with $CO_2$ capture the values of kg $CO_2$-eq per kWh obtained in this study (e.g. 0.34 kg $CO_2$-eq/kWh for MEA 30% in subcritical plant) are consistent with the value reported by Clarens et al., i.e. 0.33 kg $CO_2$ eq/kWh for MEA 30% and in the range of literature values reported in their paper (≈ 0.20 to 0.35 kg $CO_2$ eq/kWh) (Koornneef et al., 2008; Clarens et al., 2016; Viebahn et al., 2007). Table S12, presents carbon footprint for different scenarios in terms of energy penalty consideration in this study.

In terms of source of uncertainties in LCA results due to adding $CO_2$ capture unit into the plants the effects of some variables and parameters were investigated as will be described in the next sentences. The consumption amount of caustic (NaOH) in the $CO_2$ capture unit reclaimer for the MEA regeneration from heat stable salts and amine makeup replacement for the amine lost due to degradation, foaming and emission were studied, to see their effects on the carbon footprint, and other environmental impacts of the plants. The LCA presented no considerable sensitivity over NaOH and amine makeups and no noticeable change in carbon footprints. However, in terms of other categories such as acidification, eutrophication and fossil fuel depletion, especially amine makeup presents some environmental impacts (Figs. S5 and S6, Supporting Information). Nevertheless, considering the variation range of NaOH and amine makeup in this sensitivity study, their effect on the various environmental categories was relatively low. The numerical values related to Figs. S5 and S6, are listed in Tables S13 and S14.

The sensitivity of carbon and water footprints to the auxiliary electricity requirement for the unit operations was investigated. Figs. S7 and S8, Supporting Information, show that the carbon footprints (kg $CO_2$-eq/kWh) reduce as auxiliary power requirements for $CO_2$ capture unit are decreased. It can be seen, as auxiliary electricity use in the $CO_2$ capture unit decreases 50% (Aux_Power_Factor 0.5 in Fig. S7) from its nominal value (around 50 MW in this study), the carbon footprint reduces more than 10%. However, in terms of water footprint no significant change with auxiliary electric power of $CO_2$ capture unit was observed. Less or even no sensitivity of water footprint in all evaluations of $CO_2$ capture unit parameters is mainly due to huge water consumption of base plant compare to other units, which makes water consumption changes of other units unnoticeable in overall plant water footprint.

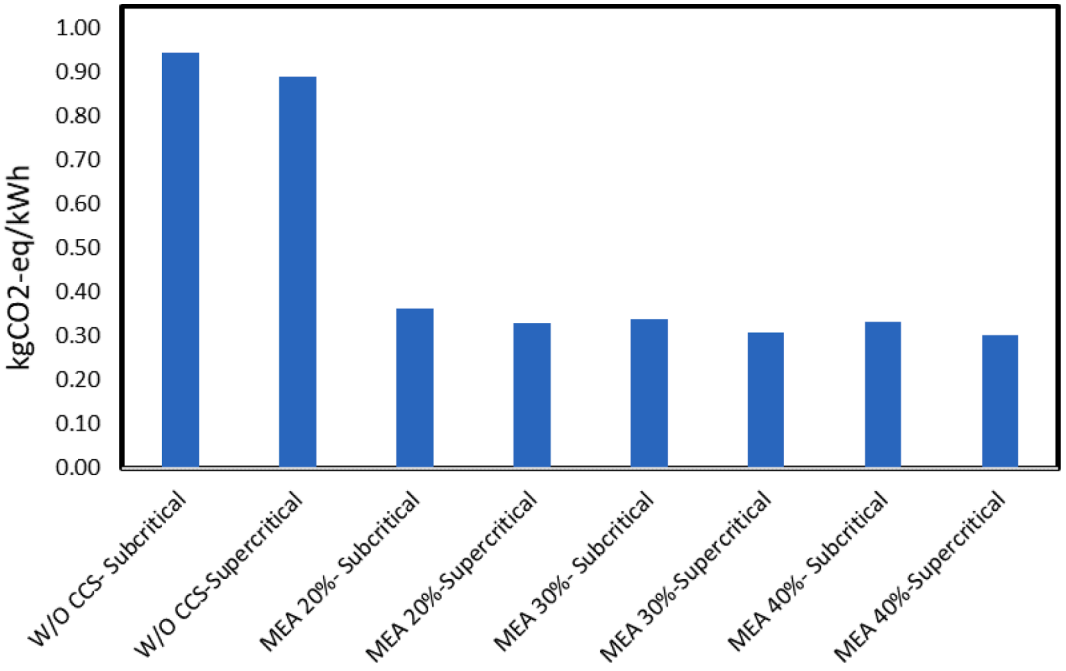


**Fig. 3.** The power plants life cycle carbon emission, carbon dioxide equivalent production per kWh energy produced for each plant, based on method of IPCC 2013 GWP 100a.

According to Fig. 4, the water footprint of the supercritical plants is lower than subcritical counterparts, which follows the same trend as the $CO_2$ equivalent production per energy produced for sub and supercritical plants displayed in Fig. 3. In terms of amine concentration, no noticeable change in water footprint is observed for different amine concentrations investigated in this work. However, it can be seen that water footprint of plants is significantly increased with the addition of a $CO_2$ capture unit. In all cases, with or without $CO_2$ capture unit, the supercritical plant presents around 10% decrease in water footprint compare to them subcritical counterparts, Table S15, Supporting Information.

The water footprint of each individual unit process in a subcritical plant with $CO_2$ capture unit and MEA concentration of 30 wt% are displayed in Fig. S9, Supporting Information. It is clear that, the $CO_2$ capture unit followed by FGD, and waste end of life have most water consumption during power plant lifetime. Since the base plant water consumption is order of magnitudes higher than other units its participation is not shown in Fig. S9.

Amine concentration changes, can affect the heat of regeneration of solvent (Abu-Zahra et al., 2007). Therefore, aqueous solutions of MEA with different concentrations of amine can provide different heat of regenerations. Fig. 5 displays the effect of the heat of regeneration of solvents on the carbon footprint, kg $CO_2$-eq/kWh, and water footprint $m^3$/kWh. As can be seen the carbon footprint is decreased from 0.36 to 0.0.33 kg $CO_2$-eq/kWh as amine concentration increases from MEA 20 wt% to MEA 40 wt%, with heat of regenerations of 5538 and 4205 kJ/kg$CO_2$, respectively. In more clear form, Fig. 6 summarizes the results of Fig. 5 and displays the trend of carbon footprint as a function of solution heat of regeneration in a converted unit (i.e. kJ/$mol_{CO2}$). Therefore, according to the LCA result and as it was expected, heat of regeneration of solvent can make a considerable change in carbon footprint of overall process, and less heat of regeneration means lower carbon footprint. On the other hand, as Fig. 5 shows, the heat of regeneration for the solutions studied in this work does not have noticeable impact on water footprint.

The $CO_2$ capture efficiency (CE) defined as, (Moles $CO_2$ in - Moles $CO_2$ out) / (Moles $CO_2$ in) (IECM Technical Documentation: Amine--based Post-Combustion $CO_2$ Capture 2018), is an important parameter in a plant performance. Fig. 7 displays that CE does not have significant impact on the water footprint, while the 10% higher CE can decrease the carbon footprint more than 20%.

Figs. 8 and 9 display the environmental impact of all plants according to the categories included in ReCiPe 2016 Midpoint (H) V1.03, and TRACI 2.1 V1.05, respectively. Table 2 displays the nomenclature used and corresponding unit on various categories graphically shown in Fig. 8. As can be seen from Figs. 8 and 9, adding the $CO_2$ capture unit to the power plants significantly decrease their carbon footprint, and has positive impact on decreasing the global warming, which is consistent with Fig. 3. For other categories such as water footprint, ozone depletion, ionizing radiation, mineral resource scarcity, marine eutrophication, freshwater eutrophication, smog, respiratory effects and fossil fuel depletion, addition of the $CO_2$ capture unit increases the environmental impact, which in some part is consistent with literature results (Clarens et al., 2016). As can be seen, in all categories, supercritical power plants display better performance compare to subcritical. The original data for Figs. 8 and 9 are given in Tables S16 and S17, Supporting Information.

Fig. 10 displays the CE effect on various environmental categories for a subcritical plant with $CO_2$ capture unit, MEA 30 wt%, through ReCiPe 2016 Midpoint (H) V1.03 method. The three different capture efficiencies of 80, 90 and 95%, were investigated for this purpose. As it is expected, higher CE has a smaller carbon footprint, while for other categories, the lower CE has lower impacts.

A sensitivity analysis over the percent of gypsum in the waste of FGD unit was performed. The results presented no meaningful ($\leq$10%) changes in kgCO2-eq/kWh. The sensitivity analysis of other environmental impacts through Recipe 2016 methods if 20% less gypsum was provided in FGD waste were investigated, which showed no meaningful differences in all categories. All changes were less than 2% variation.

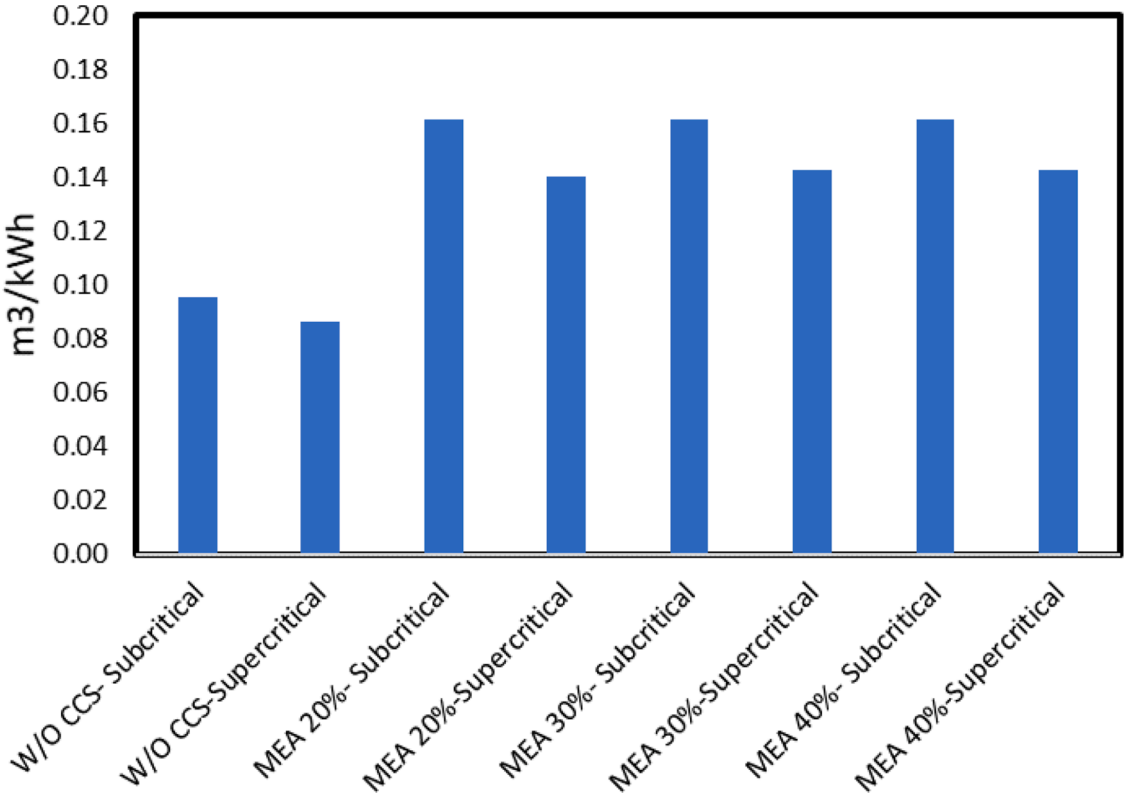


**Fig. 4.** The power plants life cycle water footprint in cubic meter water consumption per kWh produced for each plant, based on the method of AWARE V1.02.

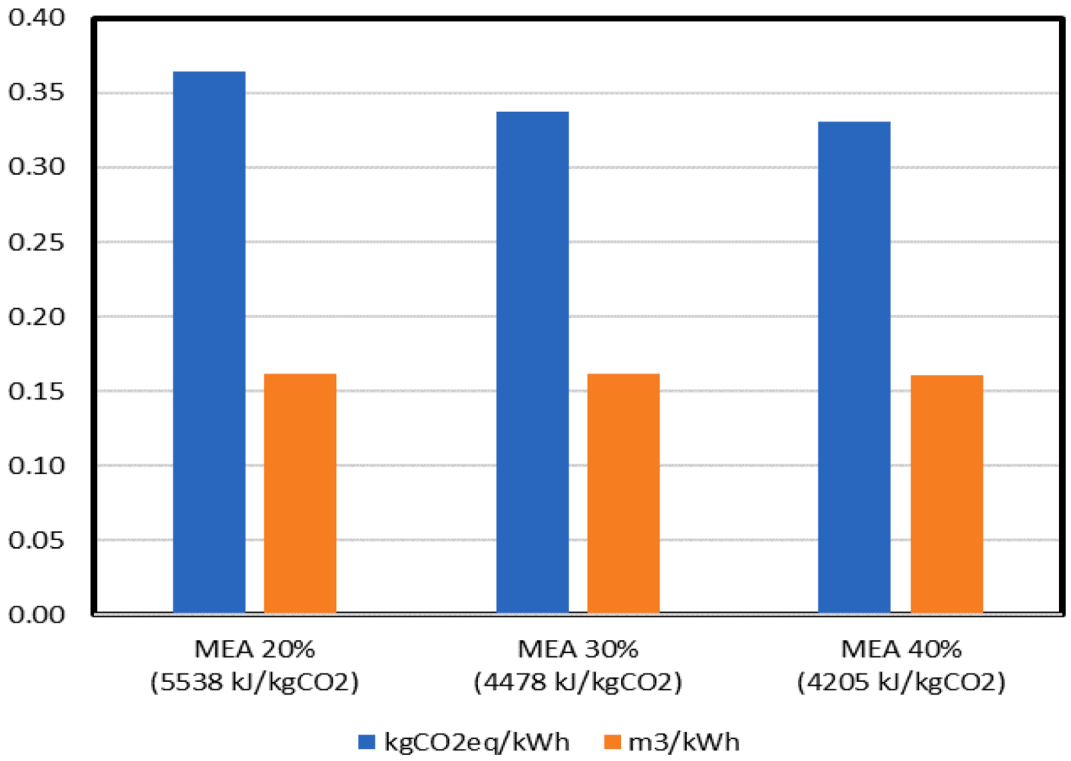


**Fig. 5.** The effect solvent heat of regeneration (and solvent concentration) on carbon and water footprints, kg $CO_2$-eq/kWh and $m^3$/kWh, respectively, in subcritical power plant.

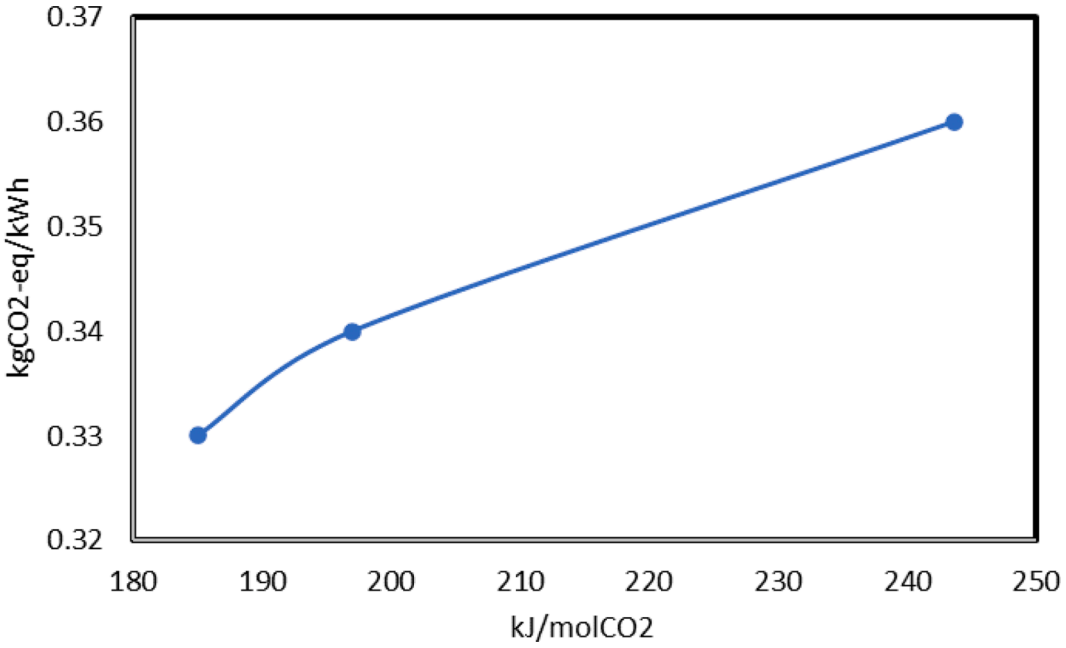


**Fig. 6.** The effect of solvent heat of regeneration (kJ/$mol_{CO2}$) on carbon footprints (kg $CO_2$-eq/kWh), in subcritical power plant.

Fig. 11 presents the amount of low-pressure steam extracted from the power plant and its equivalent electrical values to heat the loaded solvent for regeneration as a function of amine concentration in the capture unit. As can be seen, increasing the amine concentration in the solvent decreases the amount of low-pressure steam (extracted from the steam turbines) requirement in reboiler for the solvent regeneration and also $CO_2$ capture unit electrical need. However, the $CO_2$ capture unit electrical power requirement does not vary significantly with amine concentration as steam is used for the regeneration of solvent. The results show that the steam requirement for the amine regeneration can be decreased by around 30% with amine concentration increasing from 20 wt% to 40 wt%. However, considering only three amine concentrations studied in this work, the decrease in steam requirement for the solvent regeneration does not show a linear trend, and at higher amine concentrations (e.g. >30 wt%) the slope of change is not as steep as lower amine concentrations. Therefore, in terms of the energy requirement,

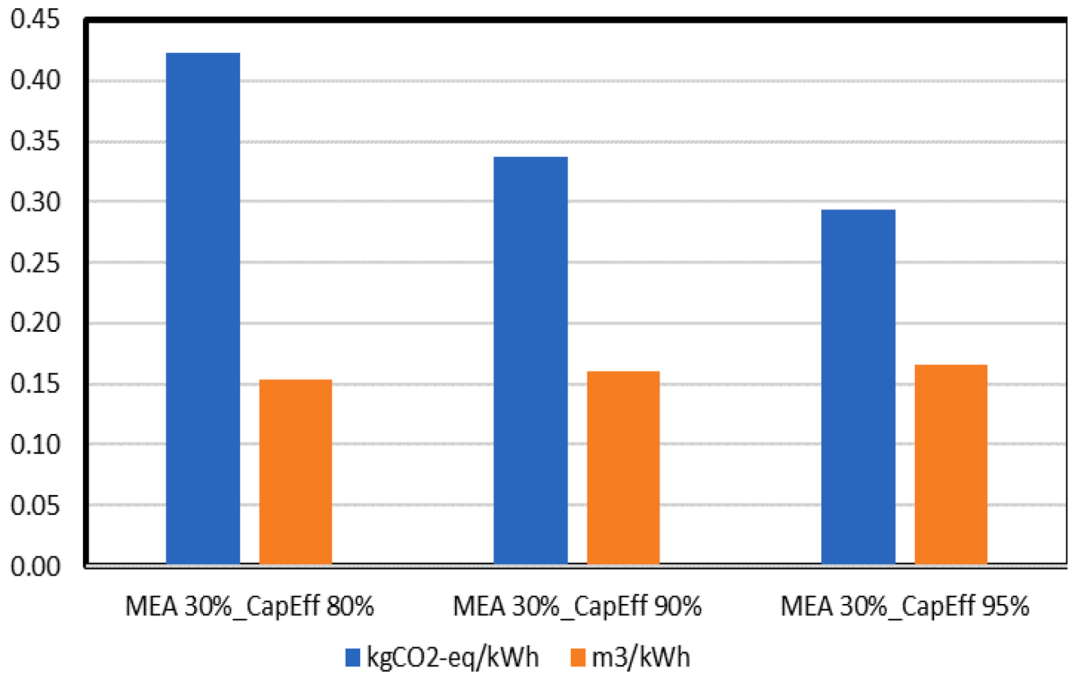


**Fig. 7.** The effect $CO_2$ capture efficiency on carbon and water footprints, kg $CO_2$-eq/kWh and $m^3$/kWh, respectively, in subcritical power plant.

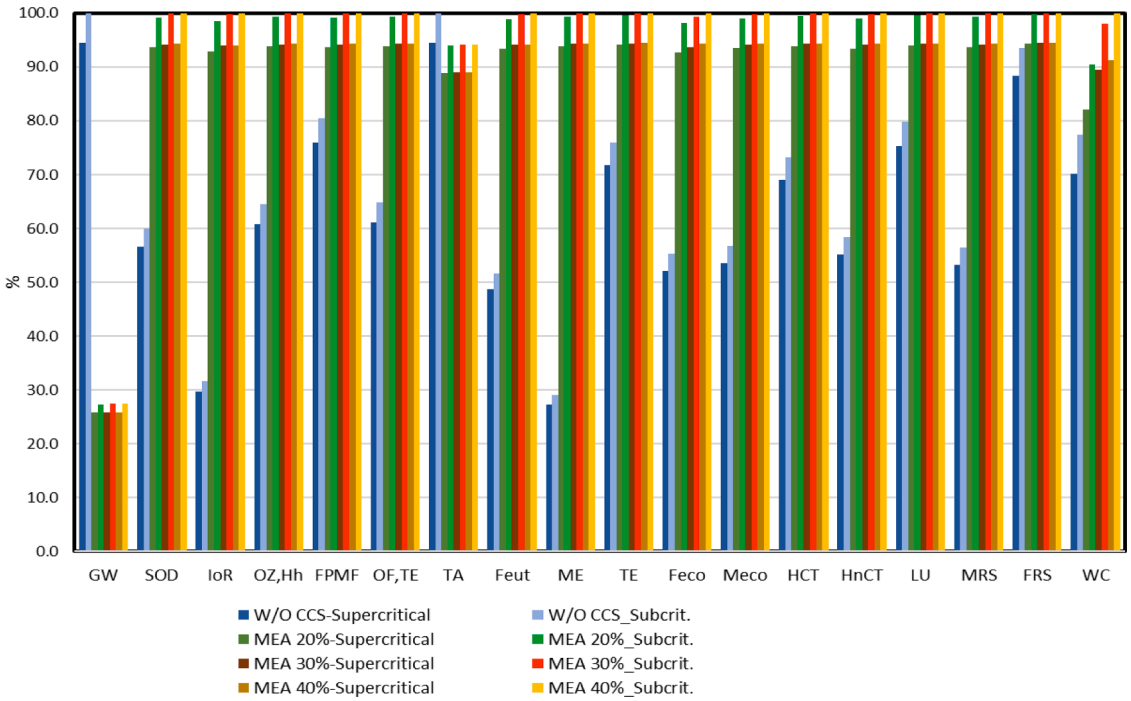


**Fig. 8.** The normalized presentation of impact assessment on various categories for the different plants according to ReCiPe 2016 Midpoint (H) V1.03 method.

increasing the MEA concentration in the solvent from 30 to 40 wt% just benefits about 6%, while the energy benefit from 20 to 30 wt% of amine concentration increasing is about 20% in $CO_2$ capture unit energy consumption.

*Ammonia-Based* $CO_2$ *Capture*: In this cyclic process (as amine-based) the flue gasses from power plant using circulating water, direct contact cooler and heat exchanger using chilled water are cooled, and most of the water in the flue gas are condensed out. The chilled flue gasses are treated in an ammonia-based solvent absorber. Then the $CO_2$ rich solvent from the bottom of the absorber is compressed to a certain pressure (e.g. 3.0 MPa in IECM model) and flows through a cross flow heat exchanger and a heater to dissolve any solids formed in the solution to the stripper at 2.8 MPa. The regenerated solvent is then returned to the absorber (Versteeg and Rubin, 2012). The ammonia based solvent for the $CO_2$ capture has low cost, higher stability in terms of solvent degradation, and needs lower energy for the solvent regeneration (Zhai et al., 2019). In order to use ammonia solution as a solvent in $CO_2$ capture process different concentrations of ammonia were suggested between 15 and 30 wt% (Darde et al., 2009; Versteeg and Rubin, 2012). In this study the 30 wt% of ammonia solvent is considered (Darde et al.,

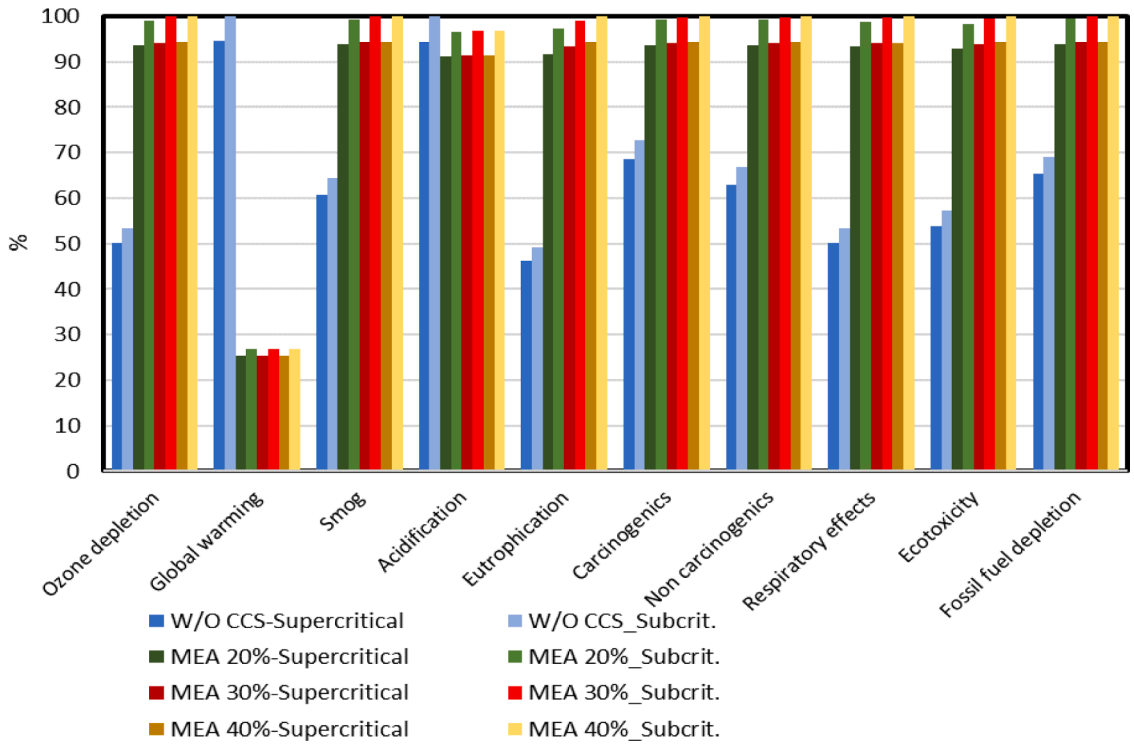


**Fig. 9.** The normalized presentation of impact assessment on various categories for the different plants according to TRACI 2.1 V1.05 method.

**Table 2**
Nomenclature used for various impact categories and corresponding units for ReCiPe 2016 Midpoint (H) V1.03 method.

| Impact category | Abbreviation | Unit |
|---|---|---|
| Global warming | GW | kg CO2 eq |
| Stratospheric ozone depletion | SOD | kg CFC11 eq |
| Ionizing radiation | IoR | kBq Co-60 eq |
| Ozone formation, Human health | OZ,Hh | kg NOx eq |
| Fine particulate matter formation | FPMF | kg PM2.5 eq |
| Ozone formation, Terrestrial ecosystems | OF,TE | kg NOx eq |
| Terrestrial acidification | TA | kg SO2 eq |
| Freshwater eutrophication | Feut | kg P eq |
| Marine eutrophication | ME | kg N eq |
| Terrestrial ecotoxicity | TE | kg 1,4-DCB |
| Freshwater ecotoxicity | Feco | kg 1,4-DCB |
| Marine ecotoxicity | Meco | kg 1,4-DCB |
| Human carcinogenic toxicity | HCT | kg 1,4-DCB |
| Human non-carcinogenic toxicity | HnCT | kg 1,4-DCB |
| Land use | LU | m2a crop eq |
| Mineral resource scarcity | MRS | kg Cu eq |
| Fossil resource scarcity | FRS | kg oil eq |
| Water consumption | WC | m3 |

2009). Figs. 12 and 13 show the carbon footprint (kg$CO_2$-eq/kWh) and water footprint ($m^3$/kWh) for a subcritical plant without and with carbon capture unit when it uses MEA-based or ammonia-based processes. As can be seen the MEA-based carbon capture unit provides better performance in terms of carbon and water footprint compare to ammonia-based. This study does not consider the thorough ammonia case for instance the effect of $NH_3$ wt% in solvent on the LCA results, and this investigation does not include other impact categories. Nevertheless, in terms of the carbon and water footprints, MEA 30 wt% presents around 15% and 30% better performance compare to the ammonia technology with $NH_3$ 30 wt%, respectively.

## 4. Conclusion

A comparative, cradle-to-gate life cycle assessment (LCA) was performed to compare the environmental impact of capturing $CO_2$ from the flue gasses in subcritical and supercritical 500 MW coal fired power plants. The geographical location for this study is in Midwest states (IA, IL, IN, KY, MI, MN, MO, ND, NE, OH, SD, WI, WV) defined in IECM software. The amine reclaimer waste composition adapted from open literature and reports, was considered in all LCA modeling in this study. The MEA concentration and capture efficiency effects on the environmental impact of different categories was investigated. The $CO_2$ capture unit was retrofitted so that the total fossil fuel (coal) input to the plants is the same for all scenarios, and no difference in the fuel input amount between plants at each subcritical and supercritical cases. However, this affects the plants electrical output while the fuel input is kept constant at 173 and 163.5 ton/hr for subcritical and supercritical power plant, respectively, Table S9.

The sensitivity of LCA results over amine and caustic (NaOH) makeups and auxiliary power used by $CO_2$ capture unit were investigated. Using IPCC 2013 GWP 100a, AWARE V1.02, ReCiPe 2016 Midpoint (H) V1.03, and TRACI 2.1 V1.05, all methods reveal that in terms of global warming, adding a $CO_2$ capture unit either amine or ammonia-based provides meaningful benefits for the environment. However, for water footprint and other impact categories such as ozone depletion, ionizing radiation, marine eutrophication, smog, and fossil fuel depletion, the plants with $CO_2$ capture unit show disadvantages compared to plants without $CO_2$ capture unit.

In terms of global warming (kg $CO_2$ -eq/kWh) and water footprint ($m^3$/kWh) the supercritical plants provide better performance compare to the corresponding subcritical ones. The results showed that in the range of MEA concentrations studied in this work, the more concentrated amine solvents provide less kg$CO_2$-eq per kWh for the plants. In terms of the water footprint, excluding the predominant water consumption by base plant, the $CO_2$ capture unit followed by FGD unit displayed the most water consumption during power plant lifetime.

The LCA evaluation shows that the MEA-based plant displays lower global warming impact and water footprint compare to the ammonia-based plant.

The additional analysis shows that, the steam used for the

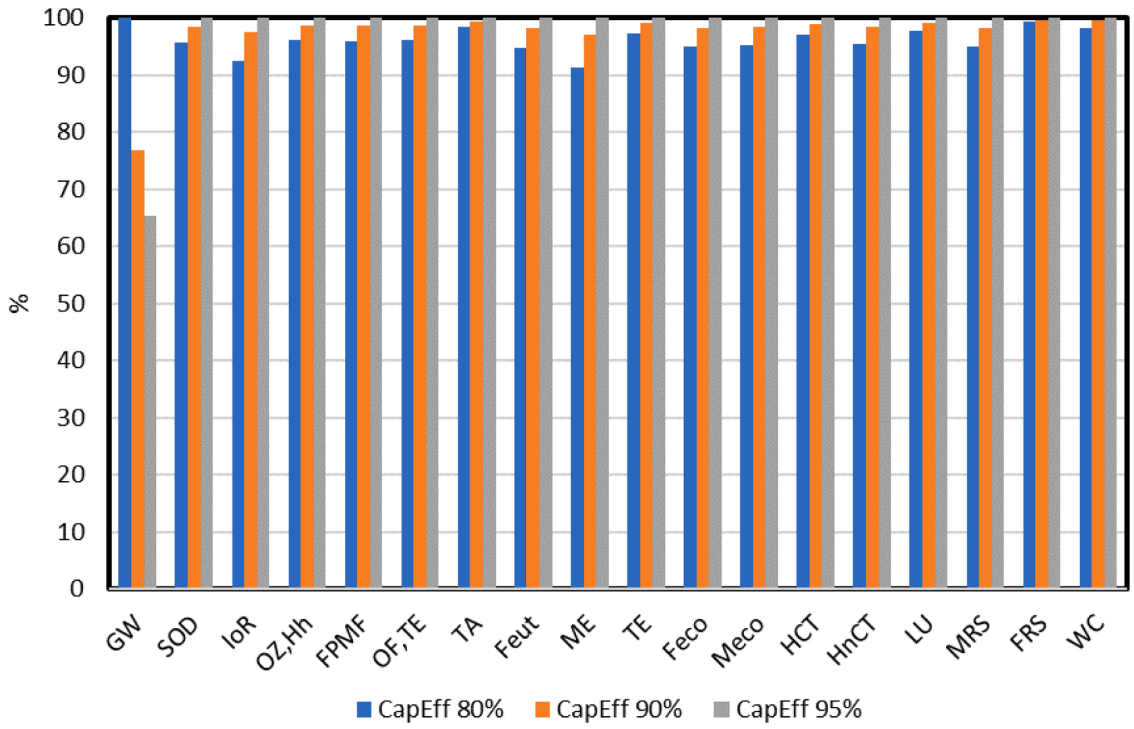


**Fig. 10.** The normalized presentation of impact assessment on various categories for a subcritical plant with $CO_2$ capture unit, MEA 30 wt% at different capture efficiencies according to ReCiPe 2016 Midpoint (H) V1.03 method.

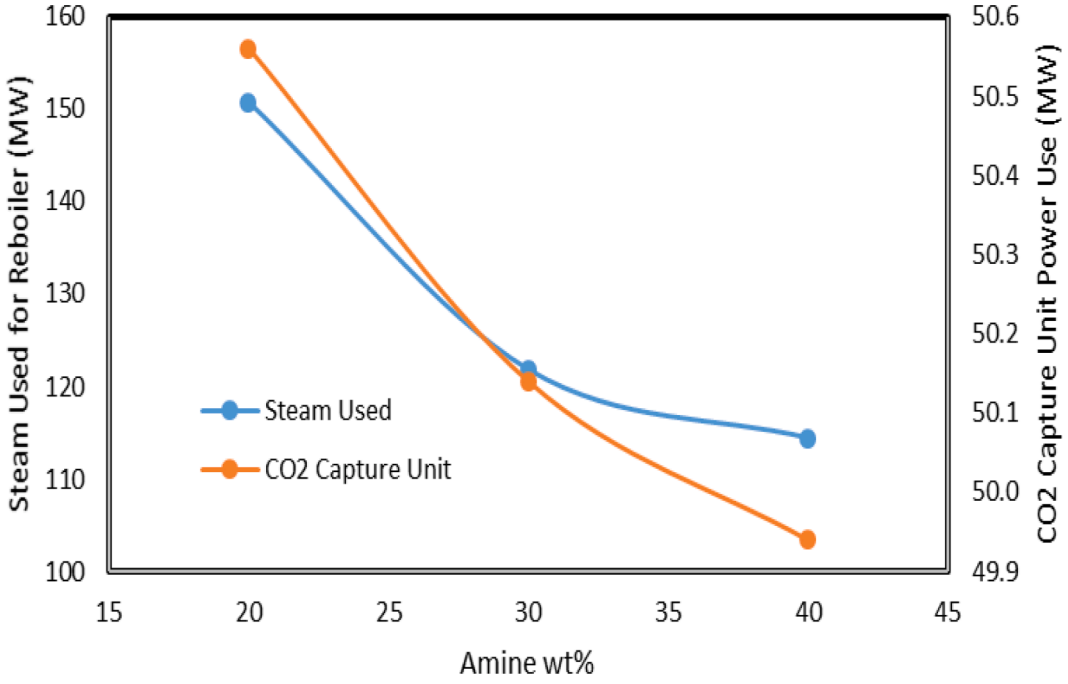


**Fig. 11.** Electrical equivalent of steam used in the reboiler for solvent regeneration and $CO_2$ capture power requirement as a function of amine (MEA) concentration in the sorbent.

regeneration of solvent can be changed significantly, and the steam requirement is decreased with increasing amine concentration. However, considering the slope of change, which is not sharp at high amine concentrations, therefore, in terms of the energy requirement (extracted steam from base plant), the MEA 30 wt% performance is close to MEA 40 wt% in $CO_2$ capture unit.

The sensitivity analysis over the percent of gypsum in the waste of FGD unit presented no meaningful ($\leq$10%) changes in $kgCO_2$-eq/kWh. Also, according to ReCiPe 2016 Midpoint (H) V1.03 methods the sensitivity of the results on the selected waste type composition (e.g. gypsum) is insignificant. In this study the data generated by IECM software were considered reliable for LCA comparative analysis. However, for more comprehensive study some selected data need to be verified by experimental data.

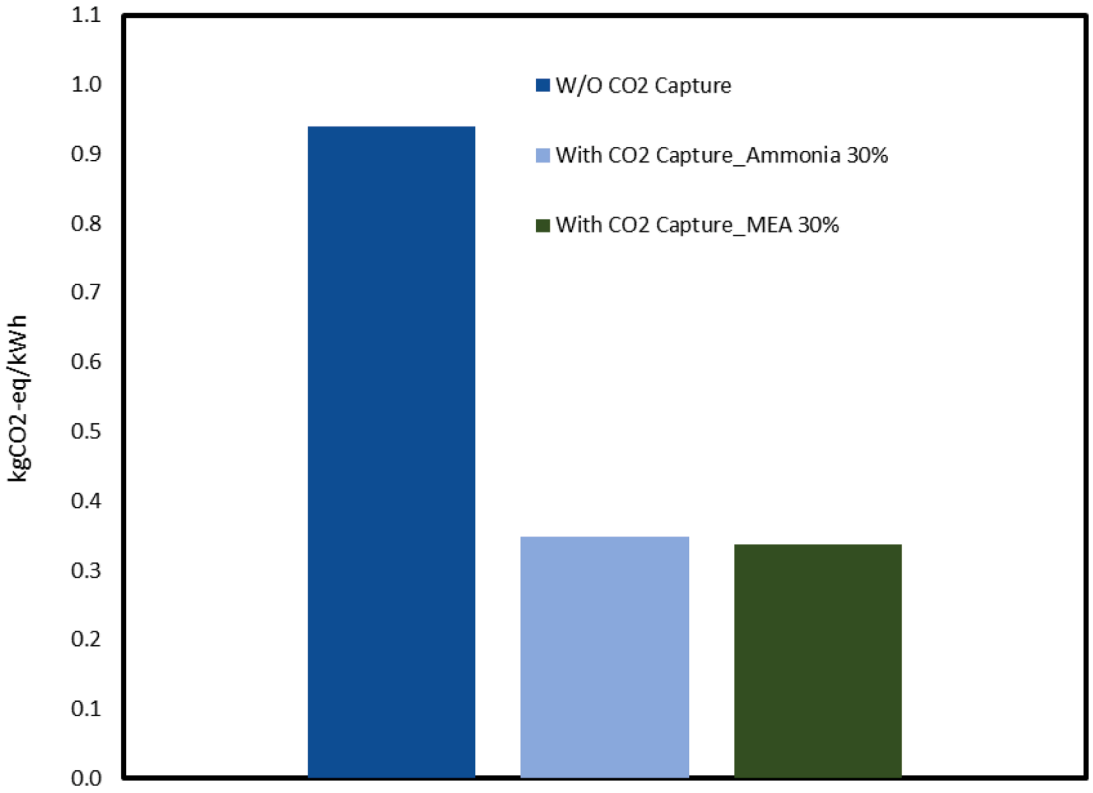


**Fig. 12.** The power plants life cycle carbon emission, carbon dioxide equivalent production per kWh energy produced for a subcritical plant without and with carbon capture unit, MEA-based vs ammonia-based processes, based on method of IPCC 2013 GWP 100a.

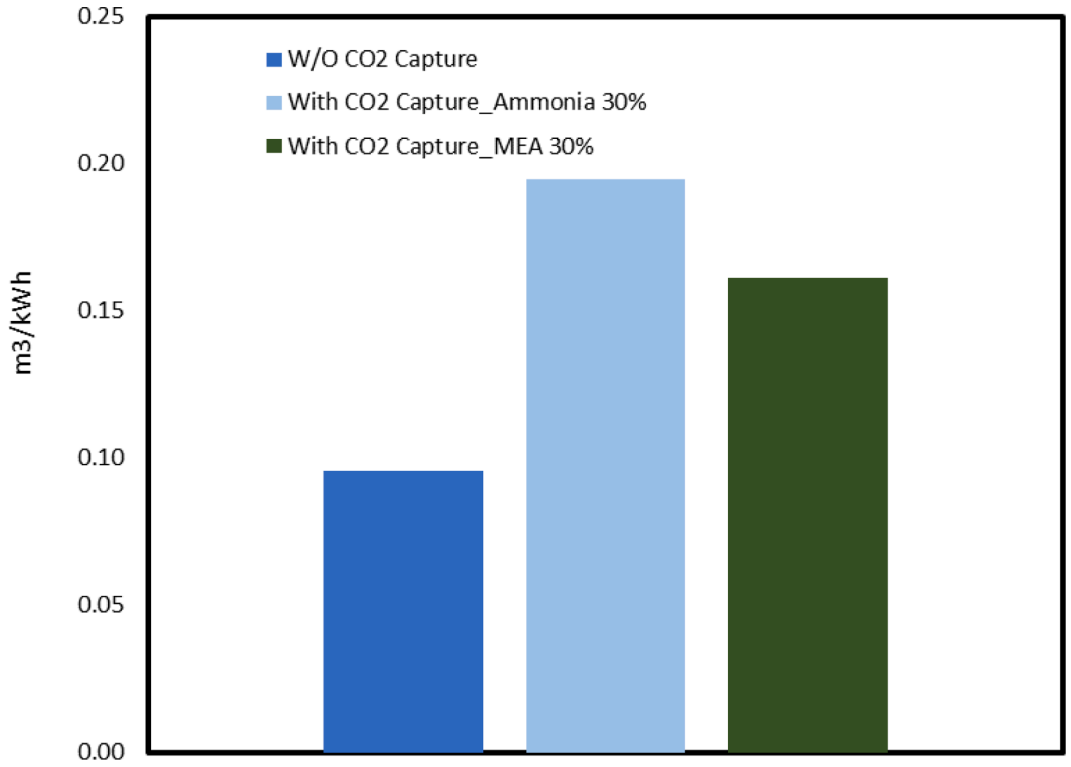


**Fig. 13.** The power plants life cycle water cubic meter consumption per kWh energy produced for a subcritical plant without and with carbon capture unit, MEA-based vs ammonia-based processes, based on AWARE V1.02, method.

## Supporting information

The Supporting Information associated with this article is available free of charge on the online version.

## 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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## Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.ijggc.2021.103535.

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