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Comparative life cycle assessment of seawater desalination technologies enhanced by graphene membranes

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HIGHLIGHTS

- Graphene oxide reduces environmental impacts of membrane distillation by 27–34 %.
- The reduction is much smaller for reverse osmosis (3–6.8 %).
- Still, that would avoid emissions of 380,000–850,000 t CO_2 eq. per year globally.
- Reverse osmosis has much lower impacts than membrane distillation.
- However, using renewable heat would make membrane distillation a better option.

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ABSTRACT

Graphene oxide (GO)-enhanced membranes are being developed to solve major limitations in both reverse osmosis (RO) and membrane distillation (MD) technologies, which include high electricity and thermal energy consumption. This study performed, for the first time, a life cycle assessment to determine the effects of using GO-enhanced membranes on the environmental impacts of seawater desalination via RO and MD. Four scenarios were evaluated and eighteen environmental impacts were quantified according to the ReCiPe impact assessment method. The average impacts for the RO-GO scenarios were lower than those of RO by 3–7 %. The reduction in the climate change impact was 3–8 %, which could avoid the release of 380–850 kt CO_2 eq. per year globally if these membranes were used in current seawater RO systems. The MD-GO scenarios had, on average, 27–34 % lower impacts than the MD scenarios. Overall, the RO-GO systems were the most favourable, with lower impacts than MD-GO for most categories. However, using solar-thermal energy instead of natural gas in MD desalination would lead to 43–93 % lower impacts in nine categories than RO powered predominantly by fossil fuels. This includes climate change, which would be 64 % lower; however, freshwater ecotoxicity would be more than four-times higher. The results of this work indicate the potential environmental benefits of GO-enhanced membranes and discuss the future developments needed to improve the performance of RO and MD.

1. Introduction

The sustainable production of potable water is one of the greatest challenges facing modern civilisation. Water accessibility is threatened mainly by the effects of climate change (droughts, glacier shrinkage and salt-water intrusion) and population growth, which result in an ever-increasing deficiency gap for many urban areas, such as Chennai (India), Amman (Jordan) and Mexico City [1–4]. Since 2016, approximately 1 % of the global population were reported to rely on desalinated water, and the United Nations predicts that this will rise to 14 % by 2025 [5]. Presently, desalination technologies generate ~95.37 million m^3 of potable water per day [6]. Reverse osmosis (RO) contributes 63 % of the market share, making it the largest, most widespread desalination technology [7]. From the total amount of desalinated water (seawater, brackish and wastewater), 34 % is produced from seawater via RO [6].

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Nomenclature

Abbreviations

AGMD	air-gap membrane distillation
CFC	chlorofluorocarbon
DB	dichlorobenzene
DMF	dimethylformamide
EOFP	photochemical ozone formation potential, ecosystem
FEP	freshwater eutrophication potential
FETP	freshwater ecotoxicity potential
FDP	fossil depletion potential
GO	graphene oxide
GWP	global warming potential
HOFP	photochemical ozone formation potential, human health
HTPc	human toxicity potential, cancer
HTPnc	human toxicity potential, non-cancer
IRP	ionising radiation potential
LCA	life cycle assessment
LDPE	low-density polyethylene
LOP	land use potential
MD	membrane distillation
MD-BAU	membrane distillation business-as-usual
MD-GOnorm	membrane distillation with GO-enhanced membranes, current situation
MD-GObest	membrane distillation with GO-enhanced membranes, best-case scenario
MDP	metal depletion potential
MEP	marine eutrophication potential
METP	marine ecotoxicity potential
MSW	municipal solid waste
ODP	ozone depletion potential
PMFP	particulate matter formation potential
PVDF	polyvinylidene fluoride
RO	reverse osmosis
RO-BAU	reverse osmosis business-as-usual
RO-GOnorm	reverse osmosis with GO-enhanced membranes, current situation
RO-GObest	reverse osmosis with GO-enhanced membranes, best-case scenario
SEC	specific thermal energy consumption
SWRO	seawater reverse osmosis
TAP	terrestrial acidification potential
TETP	terrestrial ecotoxicity potential
WDP	water depletion potential

Symbols

A	membrane area, m^2
C_{pf}	heat capacity of feed water, kJ/(kg·°C)
FFR	feed flowrate, L/h
J	membrane flux, $L/m^2 \cdot h$
ρ_f	feed water density, kg/m^3
STEC	specific energy consumption, kWh/m^3
T_{CO}	temperature at the evaporation channel inlet, °C
T_{EI}	temperature at the cooling channel outlet, °C

Seawater RO (SWRO) requires electricity to power the pumps that maintain high operating pressures of 55–70 bar [8,9]. The reported specific energy consumption (SEC) for SWRO ranges from 2 to 4.5 kWh/m^3 [10–12], which can lead to a high global warming potential (GWP) if electricity from fossil fuels is used (1.8–6.1 kg CO_2 eq./m^3 potable water produced) [13–15]. Using electricity exclusively from renewable sources could decrease the GWP by 68–90 % [16,17], but this could lead to intermittency of supply and subsequent plant shut-downs [18]. Grid-scale diversified renewable energy sources are required to overcome this issue, as well as the development of better batteries for storage [18]. Water recovery is also limited to between 30 % and 50 % [19], which consequently produces 142 million m^3 of brine per day [6]. The brine has about double the normal concentration of salts, which can reduce the amount of dissolved oxygen in seawater and lead to hypoxia in marine organisms [20,21]. Other chemicals (iron chloride and sodium hypochlorite) are also present in the brine and are toxic to marine life even at extremely low concentrations [22]. Long-term studies on brine impacts have also found decreases in the abundance and variety of species at the outfall of the brine discharge [23]. Extensive pre-treatment is required to minimise fouling and scaling of the SWRO membranes, which leads to additional energy consumption [24] and chemical usage [25]. These in turn lead to the high environmental impacts associated with SWRO (e.g. global warming potential, eutrophication and ecotoxicity), which are higher than the impacts of alternative techniques for producing potable water, such as wastewater reuse and rainwater harvesting [26].

These limitations motivate the research into alternative seawater desalination techniques [10,27–29]. Membrane distillation (MD) is one such technique, which is able to recover potable water at atmospheric pressure and reasonably low temperatures (30–90 °C) [30,31]. MD can be powered by low-grade energy, such as waste heat [32] or solar thermal [33], and can be used for a wide range of feed waters, including RO brine [34], urban water recycling [35] and textile wastewaters [36]. Nevertheless, MD is currently not widely commercialised owing to the high thermal energy requirements [37] and pore wetting [38] which affects the long-term process stability.

Nanomaterials, such as graphene and graphene oxide (GO), are currently under research to enhance membranes used for desalination purposes. The addition of GO into current polymeric MD membranes results in increased porosity, which consequently increases the flux and decreases energy consumption. The presence of oxygen-containing functional groups allows GO to disperse in water and organic solvents so it can be easily added as a nanofiller for polymer nanocomposites [39]. For RO systems, previous studies focused on GO-enhanced membranes for increasing the permeate flux [18,40]. This is because molecular modelling simulations indicated there could be significant reductions in energy consumption (of about 15 %) for SWRO [41]. However, current RO facilities have been optimised to the point where the energy consumption is near the thermodynamic minimum [42]. Therefore, at this point, flux improvements have a minor effect on the energy consumption [43]. Furthermore, increasing the permeate flux may actually exacerbate the fouling rate by increasing concentration polarisation and fouling-layer compression [44,45].

A more appropriate use of GO-enhanced membranes in RO would be to utilise their anti-fouling properties. In SWRO, fouling causes a 7 % reduction in permeate flux per year, requiring the membranes to be replaced after approximately five years [46]. The permeate flux decline is usually offset by increasing the operating pressure. Consequently, the SEC increases by approximately 8 % per year [47]. The use of GO-enhanced membranes has shown to reduce the flux decline associated with fouling during RO. Ashfaq et al. [40] found that RO membranes containing GO reduced flux decline from 22 % to 15 % over an 18-h experiment. This could reduce the RO energy consumption and the energy required for pre-treatment, while increasing the membrane lifetime [18,48,49]. However, the environmental consequences of using GO for desalination are not well understood. Additionally, it is not clear what expectations these membranes should meet in order to offset their manufacturing impacts [50]. These can be evaluated by performing a life cycle assessment (LCA), which is a widely adopted methodology for evaluating the potential environmental impacts of water treatment technologies [26].

At the time of writing, 29 LCA studies on the production of potable water via SWRO are available, of which seven include MD [15,51–55]. Furthermore, there are 27 LCA studies on graphene (and its derivatives) across a wide range of topics, including graphene synthesis [56,57], upscaling/methodological guidance [58,59], coatings [60], energy storage [61,62] and adsorbents [63]. Of these, nine involve GO, but there are only two studies on GO-enhanced membranes [50,64] and neither is on desalination. As far as the authors are aware, no studies have considered GO-enhanced RO or MD.

Despite the considerable number of desalination studies, they can give an incomplete picture of the environmental impacts, as found by Zhou et al. [65]. The authors reviewed 30 desalination LCA studies (using various technologies) and reported systemic issues arising from incomplete system boundaries. This was supported in a review by Lee et al. [29], who also found that many desalination LCA studies only analysed one or few impact categories. The review had two major recommendations: to examine emerging technologies for desalination and to pay particular attention to impacts associated with chemicals usage. This is because technologies that aim to reduce the energy consumption may be shifting the environmental burden towards the chemical usage stage (through enhanced pre-treatment). A more comprehensive environmental assessment was also recommended in the review by Lee et al. [29] which compared the LCA studies of RO against emerging technologies, such as forward osmosis, capacitive deionisation and MD. This review also highlighted the trade-off between carbon emissions and chemical treatment, whereby methods to reduce the GWP resulted in higher impacts associated with the use of chemicals. This suggests that desalination methods with a lower reliance on chemicals, i.e. lower propensity for fouling, should be explored.

Previous LCA studies on MD concluded that this option was favourable over RO when dealing with high saline feed concentrations, as energy demand is almost independent of the salt concentration [15,51,66]. However, the previous studies excluded a number of chemicals used in the membrane and module manufacturing processes that could have a significant influence on the impacts. For example, Yadav et al. [67] assessed the environmental impacts of polymeric membrane production for hollow-fibre membranes. The authors found that the use of toxic solvents was the main source of several environmental impacts, including GWP, human toxicity and fossil resource depletion. Other studies also considered solar-driven RO and MD [68,69] but many excluded significant environmental impacts, such as freshwater ecotoxicity and metal depletion, that generally are higher for solar than for fossil-fuel based energy sources [70].

To fill the abovementioned knowledge gaps, this study performed for the first time a comparative LCA of the two desalination technologies enhanced by GO membranes. Aiming to identify the environmental implications of different options, the systems operating with and without GO-enhanced membranes were considered in turn. A nanocomposite membrane was evaluated, which involves mixing GO within the polymer membrane during a well-established membrane production method for desalination purposes. The influence on the impacts of different parameters, including energy sources and consumption, were explored through sensitivity analyses. The results of this study are intended to help industry and policy makers identify environmentally sustainable options for water desalination in the near future under a range of different operating conditions and geographic locations. Moreover, the study also informs what research needs to be carried out to improve the membranes and minimise the impacts of desalination.

2. Methodology

The study followed the attributional approach according the ISO14040 and 14044 LCA standards [71], as detailed in this section. Goal and scope of the study are defined next, followed by inventory data and an overview of the impact assessment method. The interpretation of the findings can be found in Section 3.

2.1. Goal and scope definition

The main goal of this study was to estimate and compare the life cycle environmental impacts of RO and MD operated with and without GO-enhanced membranes. Although MD and RO have quite different SEC (50–200 kWh/m^3 [59] vs 2–4.5 kWh/m^3 [10–12], respectively) and use different types of energy (electricity vs heat), they are both used for water desalination. Thus, they serve an equivalent function and can be compared on the basis of the same functional unit. It is also worth comparing them to determine process improvements needed for MD to compete energetically with RO.

To achieve the goal of the study, three scenarios were evaluated for each technology (for details on the assumptions and methods used, see Section 2.2):

- Business as usual (RO-BAU, MD-BAU): RO and MD plants without GO-membranes (i.e. as they currently operate), used as a base case for benchmarking with the other scenarios;
- GO normal (RO-GOnorm, MD-GOnorm): RO and MD plants with GO-enhanced membranes based on current operating practices; and
- GO best (RO-GObest, MD-GObest): RO and MD with GO-enhanced membranes representing potential future best scenarios. (Note that a worst-case scenario was not considered because it was assumed that the GO-enhanced membranes would only be implemented if they were shown to offer a step-wise improvement.)

The functional unit was defined as '1 m^3 of produced potable water from seawater', which is a common unit for in LCA studies of desalination technologies [15,72–74]. The plant was assumed to be located in Andalucia, Spain. This region was selected as it has the largest desalination plants in Europe and also has extensive legislation in place to increase water accessibility through non-conventional resources [75]. Additionally, it is home to an MD pilot research centre at Plataforma Solar de Almería, which provided inventory data for this study. As such, the inlet seawater was modelled based on the Mediterranean Sea characteristics (salt content of 3.7 wt% and an average temperature of 20 °C). GO was also assumed to be produced in Spain as *Graphenea* is currently one of the largest global GO producers, with plans to build a 500 t/y industrial plant in Northern Spain [76].

A cradle-to-grave approach was taken, considering the following process stages: GO manufacture, membrane and module manufacture, seawater pre-treatment, treatment and waste management (Fig. 1). Some chemicals used for the manufacture of GO-membranes were omitted due to a lack of data (for details, see Section 1 in Supplementary information (SI)). Construction and dismantling of the RO and MD plants were also excluded since these have negligible impacts for larger desalination facilities [73,74,77,78]. However, depending on the background data, construction and dismantling of some other plants and equipment in the background system may be included. Electricity consumption from water abstraction and distribution were not considered because they are the same for all scenarios. Additionally, brine disposal impacts were outside the scope of this LCA, but a discussion on this can be found in Section 3.4. A 25-year lifetime was assumed for both the RO and MD plants and a useful lifetime of five years was assumed for the membranes [15,79].

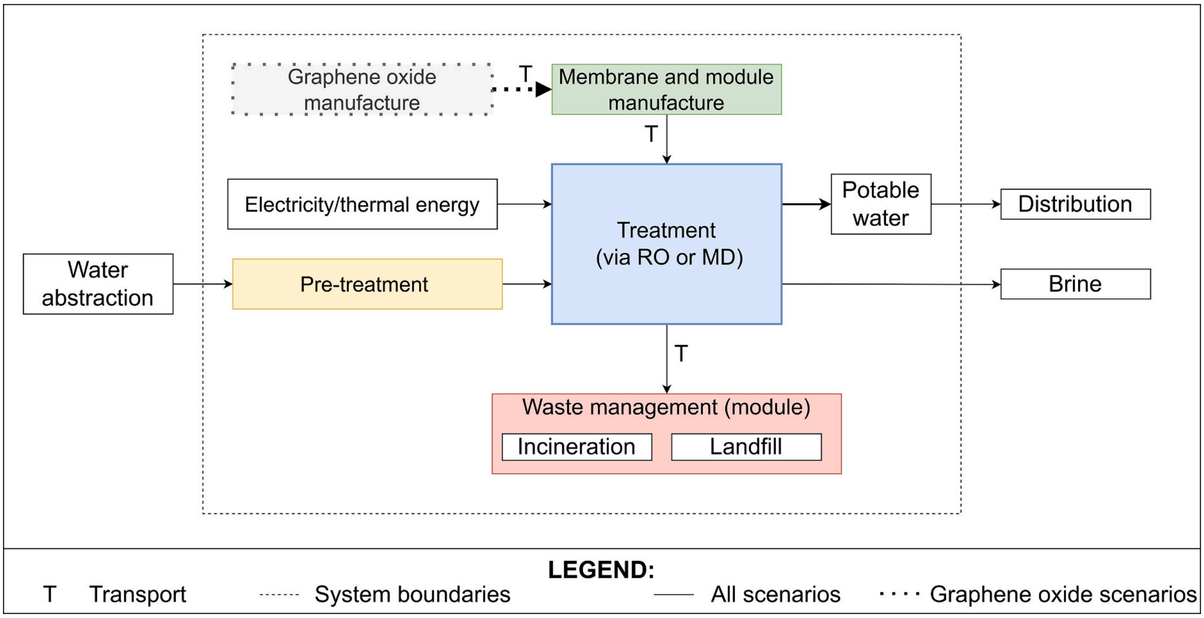

Fig. 1. A general overview of system boundaries and life cycle stages of the desalination technologies considered in the study. [Seawater abstraction, brine management and potable water distribution are outside the scope of this study. Waste management was considered only for membrane modules.]

2.2. Inventory data and assumptions

A detailed overview of the life cycle stages considered in the study can be seen in Fig. 2 and the inventory data in Table 2. Primary data were sourced from own laboratory studies and manufacturers, while secondary data were compiled from literature and the Ecoinvent v3.7 database [80]. For a full list of primary and secondary data, as well as the Ecoinvent datasets used in the modelling, see Table S1 in the SI.

The literature values for RO were taken for medium- to large-scale plants (~10,000 m^3/day) using multiple 1 m-long membrane modules

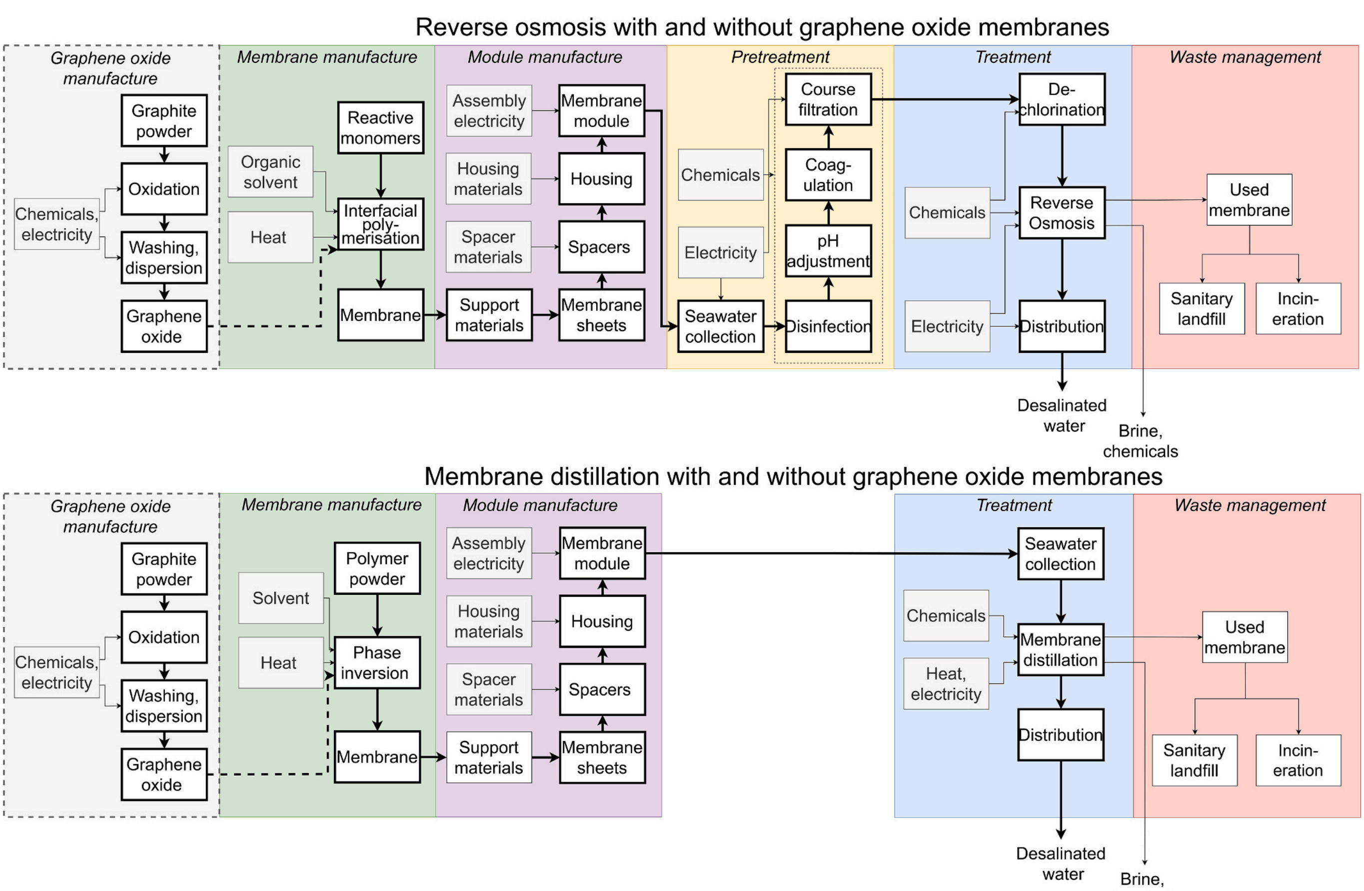

Fig. 2. A detailed overview of the life cycle stages in reverse osmosis and membrane distillation with and without graphene oxide membranes.

[81]. The membrane and module manufacture were assumed to be produced in Germany, where there are large membrane manufacturing sites. Currently, RO membranes are not routinely recycled and are treated as municipal solid waste (MSW) [82,83]. In Spain, 57 % of MSW is landfilled, 13 % is incinerated and the remaining waste is treated by other methods, such as recycling and resource recovery (e.g. biogas or fertilisers) [84]. For simplicity, recycling and resource recovery were not considered and the incineration and disposal percentages were scaled up proportionally to make up the difference (82 % landfill and 18 % incineration). The system was credited for the electricity and heat generated by incineration of RO membranes. The Spanish electricity grid for year 2020 [85] was considered in all scenarios. For the heat supply, combined heat and power and heat-only plants using natural gas in Europe were assumed, using Ecoinvent data (there were no data for Spain). The Spanish electricity mix in 2020 [85] and natural gas were considered as the avoided energy sources for the incineration credits.

The literature values for MD were sourced from a pilot-scale facility (1 m^3/day) using a 2.5 m-long membrane module [86]. The data from the pilot-scale facility are representative of a large-scale MD system because the same size modules would be used, but installed in parallel to increase the overall capacity. Aquastill is a company that specialises in manufacturing spiral wound modules for MD and the inventory data were taken from their models (see Table 1). The Aquastill manufacturing sites are based in the Netherlands which was assumed as the location for the membrane and module manufacture [87]. It was assumed that MD membranes are disposed in the same way as those discarded from RO. The following sections provide the individual process descriptions and their data sources for both RO and MD.

2.2.1. Graphene oxide manufacture

The manufacture of GO for both RO and MD was based on the modified Hummers' method [88] which involves the oxidation of graphite powder with potassium permanganate and sulphuric acid. The electricity requirements for GO production were sourced from literature [57] and the GO quantities were estimated from laboratory experiments (Table 2).

2.2.2. Reverse osmosis

RO separates all colloidal or dissolved matter from water using a semi-permeable membrane with small pores (0.1–1 nm) [89]. By size exclusion, water is able to pass through the membrane and the salts are retained in the rejected brine stream. As this process works against the osmotic pressure gradient, high hydraulic pressures of 50–70 bar [8,9] are required to generate the driving force to desalinate seawater, which accounts for most of the energy requirements.

2.2.2.1. Membrane and module manufacture. RO typically uses an ultra-thin semipermeable polyamide membrane, on top of a polysulfone support. These are built as spiral wound modules, which are composed of flat sheet membranes with spacer materials, as shown in Fig. 3. Data for their production were taken from Ecoinvent. For RO-GOnorm and RO-GObest, the GO is bound onto the polyamide surface through covalent binding as outlined in previous work [90]. This method is used to create a graphene oxide functionalised surface for antifouling purposes [40,91]. For further details on the membranes and module manufacture, see Section 1 in the SI.

Table 1
Life cycle inventory for the manufacture of graphene oxide (per 1 m^3 of potable water produced from seawater).

Input	RO-GOnorm & RO-GObest	MD-GOnorm & MD-GObest	Reference
Graphite powder, mg	0.29	0.86	[88]
Potassium nitrate, mg	0.26	0.77	[88]
Sulphuric acid, mL	0.33	0.97	[88]
Potassium permanganate, mg	1.3	3.9	[88]
Hydrogen peroxide, μL	0.88	2.6	[88]
Electricity, kWh/m^3	0.51	5.4	[88]

2.2.2.2. Pre-treatment. The pre-treatment stage was modelled based on a conventional system, which involves disinfection with chloride, pH adjustment, coagulation/flocculation with iron chloride and granular and cartridge filtration to remove larger solids. Data on the chemicals and required electrical energy were gathered from literature [10,98,101]. Data were taken only for existing medium-to-large SWRO systems with conventional pre-treatment techniques. In the cases where there were multiple reported values for similar systems, the average was assumed (Table 2).

2.2.2.3. Treatment. Seawater treatment comprises dechlorination, the RO process and distribution of water. The chorine is removed using sodium bisulfite, which prevents membrane oxidation. Sodium hypochlorite is used as a clean-in-place (CIP) chemical in the RO process [97].

The electricity demand for RO is reported in the range of 2 and 4.5 kWh/m^3 for modern (<10 years old), single pass, medium-to-large plants (~20,000 m^3/day) [12,13,101]. The lower range refers to newer and more energy-efficient plants; however, this can lead to higher energy consumption in the pre-treatment stage [18]. Different feed salinities, target conditions and equipment efficiency can also lead to a range of energy-consumption values [102]. To minimise uncertainty, the upper and lower ranges were considered, with the midpoint of 3.25 kWh/m^3 assumed in the base case.

The water recovery was assumed to be 40 %, which is between the 30–50 % reported range [6]. The average flux decline due to fouling was 7 % per year for five years [46]. The impact of reduced flux on the energy consumption was modelled using the reverse osmosis system analysis (ROSA) software from DuPont Water Solutions [103]. It was found that the energy consumption increased by 25 % over the five-year module lifetime. It was assumed that the literature reported range (2–4.5 kWh/m^3) did not account for the increase in energy consumption due to membrane fouling. Assuming the energy consumption increases linearly each year, the average increase over the five-year module lifetime is 12.5 %. Thus, the electricity requirements for the base-case RO scenario were scaled up to 2.25–5.06 kWh/m^3. For the RO-GOnorm scenario, it was assumed that GO could reduce the flux decline (according to current anti-fouling studies [40,104]) such that the increase in energy consumption is 14 % higher at the end of the module lifetime (with an average increase of 7 % over five years). Thus, the electricity requirements are between 2.14 and 4.82 kWh/m^3. For RO-GObest, the energy demand was 2–4.5 kWh/m^3, assuming no flux decline in the best case and thus no additional energy consumption [40].

2.2.3. Membrane distillation

MD works by exploiting the difference in vapour pressure between the water and dissolved salts [18]. In MD, the seawater is heated to around 80 °C and is tangentially circulated along the surface of a hydrophobic membrane, where water vaporises and is able to pass through the membrane and condense on the other side. Different MD configurations exist and, in this study, air-gap (AGMD) was considered for its commercial applications [86,100,105]. In AGMD, the water vapour passes across an air-gap until it contacts a condenser plate that is kept cool by a separated cooling stream. The cooling stream is normally incoming pre-heated seawater, such that the latent heat of condensation can be recovered to improve the process heat efficiency. The non-volatile salts, as well as liquid water, are retained and form the waste brine solution. The vapour pressure gradient is maintained by a concurrent heating and cooling system [18].

2.2.3.1. Membrane manufacture and module assembly. The membrane manufacture was based on the polymerisation of fossil-derived ethylene

Table 2
Inventory data for reverse osmosis (RO) and membrane distillation (MD) and respective GO-enhanced membranes (per 1 m^3 of potable water produced from seawater).

Stage	Inputs, outputs and activities	BAU		GOnormal		GObest		Comment	Reference
		RO	MD	RO	MD	RO	MD		
Membrane manufacture	Polyethylene powder, g	–	0.18	–	–	–	–	Membrane material	[92]
	Liquid paraffin, g	–	0.054	–	–	–	–	Liquid lubricant for polymerisation	[92]
	Polyvinylidenfluoride, g	–	–	–	0.18[a,b]	–	0.18[a,b]	Membrane material	[93]
	Dimethylformamide, g	–	–	–	0.12[a,b]	–	0.12[a,b]	Solvent for phase inversion	[93]
	Deionised water, L	–	–	–	0.12[a,b]	–	0.12[a,b]	Non-solvent for phase inversion	[93,94]
	Graphene oxide, mg	–	–	0.18[a,b]	0.54[a,b]	0.18[a]	0.54[a,b]	Amount of GO required	[93,95]
	Heat, kWh	–	0.0029	–	0.0029	–	0.0029	Heat required during manufacture	[92,94]
	RO membrane module, cm^2	9.1	–	9.1	–	9.1	–	Ecoinvent data on “Market for seawater reverse osmosis module”[c]	[96]
	Polypropylene, g	–	0.63[a]	–	0.63[a]	–	0.63[a]	Spacer and housing material	[15,94]
	Polyethylene terephthalate, g	–	0.28	–	0.28	–	0.28	RO spacer material and MD condenser material	[94]
	Aluminium, g	–	0.28	–	0.28	–	0.28	MD condenser material	[86]
	Coated steel, g	–	0.89	–	0.89	–	0.89	MD housing material	[94]
	Electricity, kWh	–	0.0036	–	0.0036	–	0.0036	Electricity required for rolling process	[94]
	Transport, km			2400	2200	2400	2200	Lorry, 7.5–16 t, EURO6 from Spain to Germany/Netherlands (GO)	[96]
	Transport, km	2400	2200	2400	2200	2400	2200	Lorry, 7.5–16 t, EURO6 from Germany/Netherlands to Andalucía	[96]
Pre-treatment	Seawater, m^3	2.5	20	2.5	20	2.5	20	Salinity of 37 g/L	
	Sodium tripolyphosphate, g	2.6[a]	–	2.6[a]	–	2.6[a]	–	Anti-scalant	[15,97]
	Ferric chloride, g	7.5	–	7.5	–	7.5	–	Flocculant	[97]
	Chlorine, g	7.5	–	7.5	–	7.5	–	Disinfectant	[97]
	Sodium bisulphite, g	15	–	15	–	15	–	Dechlorination	[97]
	Activated carbon, g	4.47	–	4.47	–	4.47	–	Filtration	[74]
	Sulphuric acid, g	62.5	–	62.5	–	62.5	–	pH adjustment	[97]
	Electrical energy, kWh	0.43[a]	–	0.43[a]	–	0.43[a]	–	Electricity for pre-treatment	[10,16,98,99]
Treatment	Sodium hydroxide, g	–	0.056	–	0.056	–	0.056	Clean-in-place	[15]
	Sodium hypochlorite, mg	7.5	0.81	7.5	0.81	7.5	0.81	Clean-in-place	[15,97]
	Deionised water, g	–	0.056	–	0.056	–	0.056	Clean-in-place	[15]
	Heat, kWh	–	50–200[d]	–	33–134[d]	–	33–134[d]	For desalination	[86]
	Electricity, kWh	2.25–5.06[d]	–	2.14–4.82[d]	–	2.0–4.50[d]	–	For desalination	[7,10]
Waste and emissions	Steel, g	–	0.28	–	0.28	–	0.28	From RO and MD modules. 19 % of waste incinerated and 81 % landfilled	[6,84,100]
	Polyvinylchloride, g	–	0.18	–	0.18	–	0.18		[84]
	Polyethylene terephthalate, g	0.030	0.28	0.030	0.28	0.030	0.28		
	Polypropylene, g	0.023	0.63	0.023	0.63	0.023	0.63		
	Plastic[e], g	0.012	–	0.012	–	0.012	–		
	Aluminium, g	–	0.89	–	0.89	–	0.89		
	Activated carbon, g	4.47	–	4.47	–	4.47	–	Landfilled	
	Sodium tripolyphosphate, g	2.6	–	2.6	–	2.6	–	Emissions to water (mixed and discharged with brine)	
	Ferric chloride, g	7.5	–	7.5	–	7.5	–		
	Sodium bisulphite, g	15	–	15	–	15	–		
	Sulphuric acid, g	62.5	–	62.5	–	62.5	–		
	Sodium hydroxide, g	–	0.056	–	0.056	–	0.056		
	Sodium hypochlorite, mg	7.5	0.81	7.5	0.81	7.5	0.81		
	Brine[f], m^3	1.5[a]	19[a]	1.5[a]	19[a]	1.5[a]	19[a]	Assuming recovery of 40 % for RO and 5 % for MD.	

[a] Average data.
[b] The references listed in the last column refer to the studies on the membrane manufacturing method, which was replicated in the current study in the laboratory to acquire the inventory data.
[c] Incorporates the materials shown in Fig. 3.
[d] Considered as ranges.
[e] Membrane active layer, plastic end caps and housing modelled as general plastic waste due to a lack of data.
[f] For the composition of brine, see Table S1. Impacts of brine discharged into the sea not considered.

to make low-density polyethylene (LDPE), which is a common method to produce hydrophobic membranes for MD [106]. For the scenarios with GO-enhanced membranes, membrane manufacture was based on phase inversion of PVDF using dimethylformamide (DMF), according to laboratory data and the work by Leaper et al. [93].

The synthesised membranes get assembled into spiral wound modules during module manufacture. Spacers and support materials are used and details on the materials were sourced from literature [33]; the weight contributions were estimated using RO autopsy data [94] and a previous MD LCA study [15]. It was assumed that MD roller machines

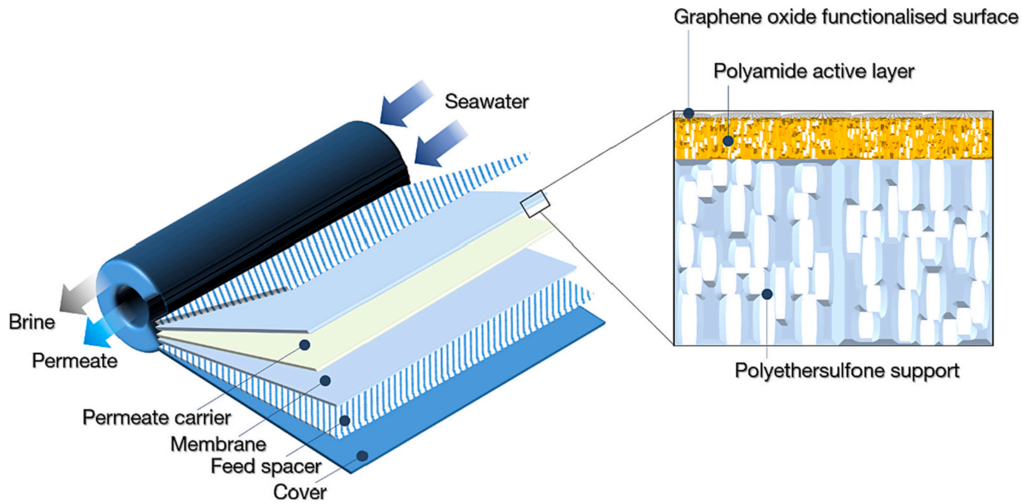

Fig. 3. Schematic diagram of a reverse osmosis membrane and module, showing the position of the graphene oxide layer (inset). Adapted from [18].

use the same amount of energy as RO machines when producing spiral wound modules. More details on this can be found in Section 1 in the SI.

2.2.3.2. Pre-treatment. MD has a higher resistance to fouling when compared with RO and it can tolerate a higher amount of suspended solids and dissolved solutes in the feed water. Thus, the pre-treatment methods are expected to be less chemically and energy intensive than RO [15,31]. There are no standard pre-treatment strategies for MD, but some are currently being investigated [107]. Current large-scale MD plants typically operate with un-treated seawater, thus it was assumed that no pre-treatment was required [66].

2.2.3.3. Treatment. The treatment stage consists of the seawater collection, membrane distillation and the distribution of potable water. Thermal energy is used to heat up the seawater prior to the desalination. Recent reported literature values for equivalent AGMD modules vary from 50 to 200 kWh/m^3 at pilot scale [86,108,109]; these ranges were considered with the middle value of 125 kWh/m^3 (Table 2). The wide range of values is attributed to recent optimisation advances and uncertainties in process operation. Data on the energy, water recovery and quantities of cleaning chemicals were sourced from pilot-scale AGMD studies [86,110,111] as data for large-scale plants were not available. For the GOnorm and GObest scenarios, the increase in permeate flux reduces the specific thermal energy consumption according to the following relationship [86]:

$$STEC = \frac{FFR \cdot \rho_f \cdot C_{pf} \cdot (T_{EI} - T_{CO})}{3.6 \times 10^3 \cdot J \cdot A} \quad (1)$$

where:

$STEC$ specific thermal energy consumption, kWh/m^3
FFR feed flow rate, L/h
ρ_f density of the feed, kg/m^3
C_{pf} heat capacity of the feed, kJ/kg°C
T_{EI} temperature at the evaporation channel inlet, °C
T_{CO} temperature at the cooling channel outlet, °C
J permeate flux, L/m^{2}h
A membrane area, m^2.

Literature for MD with GO-enhanced membranes reports a 50–74 % flux increase when compared with pristine/commercial membranes [93]. According to Eq. (1), this would result in STEC of 33–134 kWh/m^3 for GOnorm and 29–118 kWh/m^3 for GObest, respectively. Electrical energy is required for pumping and recirculation in the module and the Aquastill manufacturers estimate[1] that this would consume 0.41 kWh/m^3.

2.3. Impact assessment

The LCA modelling was carried out in Gabi v9.2 [112]. The environmental impacts were estimated according to the ReCiPe 2016 method, considering all 18 impact categories [113], as follows: climate change potential (CCP); fossil fuel depletion potential (FDP); metal depletion potential (MDP); human toxicity, cancer (HTPc); ionising radiation (IRP); photochemical ozone formation, ecosystem (EOFP); photochemical ozone formation, human health (HOFP); stratospheric ozone depletion (ODP); chlorofluorocarbon (CFC); terrestrial acidification (TAP); particulate matter formation (PMFP); water depletion potential (WDP); freshwater ecotoxicity (FETP); marine ecotoxicity (METP); terrestrial ecotoxicity (TETP); freshwater eutrophication (FEP); marine eutrophication (MEP); human toxicity, non-cancer (HTPnc) and land use (LOP).

2.4. Sensitivity analysis

A sensitivity analysis was carried out for different electricity sources to consider the effect on the impacts of situating the desalination plants in other parts of the world. Previous RO desalination studies have suggested that switching from fossil (e.g. natural gas) to renewable electricity sources (e.g. wind) can lead to a 90 % reduction in impacts across most categories [16,17]. However, using only renewables can lead to an intermittent supply of energy, which can cause shut-downs. Thus, in this study different electricity mixes were evaluated, to consider both fossil fuels and renewables, as shown in Table 3. Spain, California, United Arab Emirates and Saudi Arabia were selected for their varied electricity mixes and high reliance on desalination [114].

A further sensitivity analysis was carried out to consider the effect on the results of the following heat sources: a natural gas boiler [117], biogas from biowaste and sewage sludge burned in a gas engine [118], solar thermal [119] and waste heat from an incineration plant [120]. For this analysis, the Spanish electricity mix was kept in all cases.

[1] Personal communication (2022).

Table 3
Electricity mix for the countries/regions considered in the sensitivity analysis (percentage contribution by source for the year 2020).

Electricity source	Spain [85]	California, United States [115]	United Arab Emirates [115,116]	Saudi Arabia [115,116]
Hydroelectric	13	7.9	0	0
Wind	22	5.0	0	<0.01
Solar thermal	1.8	0.8	3.8	0.3
Solar PV	6.0	10	0.3	0
Biomass/ biogas	1.8	2.1	0	0
Geothermal	0	4.2	0	0
Nuclear	22	6.0	1.2	0
Coal	2.0	0.1	0	0
Natural gas	29	34	95	61
Other	1.1	0.1	0	0
Oil	1.0	0.01	0	39
Imports	1.3	30	0.004	0

3. Results and discussion

An overview of the results can be seen in Fig. 4 from which it can be inferred that using GO-enhanced membranes in RO would lead to an average reduction in the impacts of 3 % for RO-GOnorm and 6.8 % for RO-GObest, relative to RO without GO. For MD, the effect of GO membranes is much more pronounced, reducing the impacts on average by 27 % and 34 % for the GOnorm and GObest scenarios.

The membrane manufacturing with GO (the GOnorm and GObest scenarios) has marginally higher impacts than that without it; however, in the overall system, this is largely offset by the improved desalination performance, which reduces the impacts of treatment.

RO-BAU is the best option in 14 of the impacts when compared with MD-BAU, including climate change and marine ecotoxicity. Even when compared with MD-GObest, RO-BAU is the best option in 11 of the impacts. However, the error bars for MD are large – when comparing the lowest range for RO-GObest and MD-GObest, it can be seen that the former achieves the lowest impacts for nine categories and MD-GObest has the lowest impacts for the other nine.

Figs. 5 and 6 show the impact contributions from different life cycle stages for RO and MD, respectively. The treatment stage is the highest contributor in all impact categories for MD and in all but one category (metal depletion potential) for RO. The CIP chemicals contribute <5 % across all impacts for both RO and MD, which is small in comparison to the impacts from energy consumption during treatment and the chemicals and energy used for pre-treatment. The total contribution of waste management, transport and GO manufacture is even smaller (<2 % in RO and <2.5 % in MD), with the latter contributing only 0.02–0.59 %. These results are discussed in more detail for each impact in turn in the next sections, referring to the mean values, if not specified otherwise. For the impact values and their ranges, see Table S2 in the SI.

3.1.1. Climate change potential (CCP)

As shown in Fig. 4, the RO treatment has significantly lower CCP (83–97 %) than MD across the options considered. This is mainly due to the lower energy requirements for desalination (3.25 vs 125 kWh/m^3). For the RO-BAU and MD-BAU options, the CCP was estimated at 1 and 23 kg CO_2 eq./m^3, respectively. As MD is an emerging technology, there is a wide variation in results (9.3–37 kg CO_2 eq./m^3) owing to the differences in reported heat consumption (50–200 kWh/m^3). The variation in results is much lower for the more established RO-BAU (0.7–1.2 kg CO_2 eq./m^3).

The treatment stage has the largest contribution to CCP (Figs. 5 and 6), specifically the electricity used for RO and the natural-gas heat for MD. All other processes contribute <1 % for MD-BAU. For RO, other major contributions to the impact are the pre-treatment (15 %) and membrane/module manufacturing stages (8.9 %). The release of volatile solvents (ethane, 1.1.2-trichloro-1,2,2-trifluoro and CFC-113) to the atmosphere is the main contributor (97 %) during the membrane manufacture, while the electricity consumption is accountable for 51 % of the impact from pre-treatment.

The use of GO-enhanced membranes leads to a reduction in the energy consumption in the treatment stage, as a result of reducing fouling in RO and increased permeation flux in MD. As the treatment stage is the highest contributor in all environmental impact categories, this is reflected in a reduction in CCP of 47 % for MD-GOnorm and 54 % for MD-GObest, compared to MD-BAU (Fig. 4). However, the benefit of GO-membranes is much lower in RO, with the CCP of RO-GOnorm and RO-GObest being lower by only 3.6–8.2 % relative to RO-BAU. The reason for this is that the GO-enhanced membranes reduce the energy consumption by 5–11 % for RO and 33–41 % for MD.

The scenario that achieves the lowest CCP (0.65 kg CO_2 eq./m^3) is RO-GObest where GO-enhanced membranes prevent flux decline completely. By comparison, the minimum values for MD-GOnorm and MD-GObest are 9.9 and 5.5 kg CO_2 eq./m^3, respectively, which are still significantly higher than the maximum value for RO (1.2 kg CO_2 eq./m^3). These results show that, when MD relies on non-renewable energy, it is not competitive with RO, regardless of the use of GO-enhanced membranes. The results also suggest that, if all current SWRO plants (32.4 Mm^3/day of potable water) were fitted with GO-enhanced membranes, they would avoid emissions of 380,000–850,000 t CO_2 eq. per year. This is equivalent to the amount of greenhouse gases released by approximately 80,000–180,000 petrol cars over one year [121].

3.1.2. Resource depletion potential – fossil fuels (FDP) and metals (MDP)

MD-BAU has the highest fossil fuel depletion potential: 9.3 compared to 0.62 kg oil eq./m^3 for RO-BAU; this is due to the use of natural gas for heat supply. Metal depletion is also the highest for MD-BAU (7.2 vs 1.7 g Cu eq./m^3 for RO-BAU). This impact is largely due to the consumption of stainless steel for construction of the natural-gas plant, supplying heat for MD.

The use of GO-enhanced membranes reduces FDP and MDP by 3 % for RO-GOnorm and 6–7 % for RO-GObest. For MD, FDP reduces by a greater percentage (33 % for MD-GOnorm and 41 % for MD-GObest) because the GO-enhanced membranes lower the total energy consumption by a greater amount. A similar trend can be seen with MDP results: the use of GO-enhanced membranes reduces the impacts by 32 % and 39 % for MD-GOnorm and MD-GObest, respectively. The contribution of GO manufacture to both impacts is negligible in both RO and MD.

Overall, all RO options are much better for both impacts than any MD option with no overlaps in ranges for FDP and only a slight overlap for MDP between the lower range for MD-GOnorm and MD-GObest and upper range for the RO scenarios.

3.1.3. Human toxicity potential – cancer (HTPc) and non-cancer (HTPnc)

For HTPc, RO-BAU is better than MD-BAU by a factor of four (0.59 and 2.6 g 1,4-DB eq./m^3, respectively). The higher impact for MD-BAU is mainly due to the release of benzene during the production of natural gas. The lowest MD result in this category was estimated for MD-GObest (0.6 kg 1,4-DB eq./m^3); however, this impact is still slightly higher than that of RO-BAU (0.59 g 1,4-DB eq./m^3).

For the HTPnc, the results for RO-BAU and MD-BAU are closer than for HTPc (0.88 and 0.72 kg 1,4-DB eq./m^3, respectively). All of the MD options are better in this category than the RO alternatives, with the impact of MD-GObest being almost half that of RO-GObest (0.45 vs 0.85 kg 1,4-DB eq./m^3).

3.1.4. Ionizing radiation potential (IRP)

MD-BAU is a better option for this impact than RO-BAU (0.17 vs 0.89

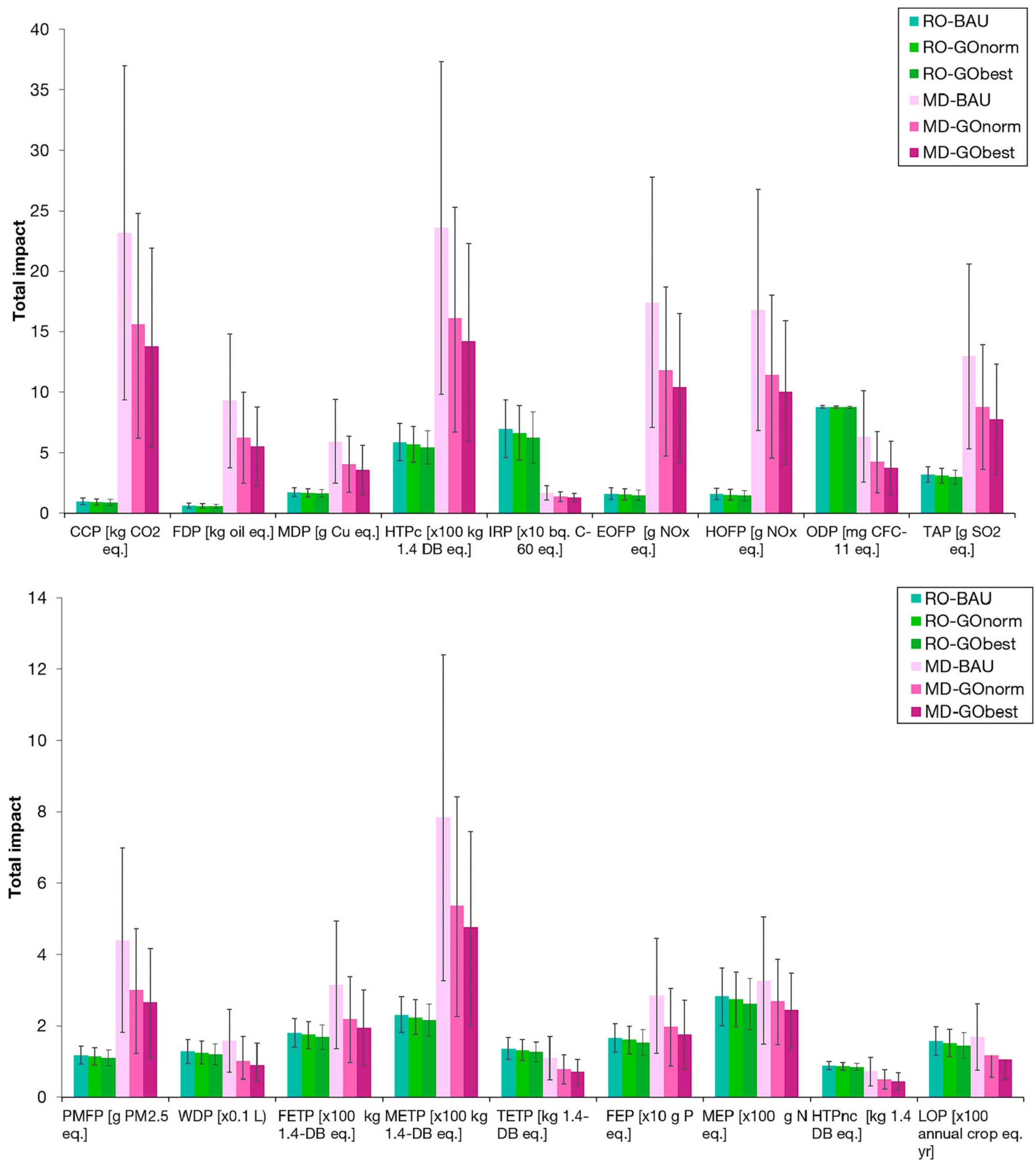

Fig. 4. Life cycle environmental impacts of different scenarios for reverse osmosis and membrane distillation. [The impacts are expressed per 1 m^3 of potable water and are calculated using the mean values in Table 1; the error bars present the minimum and maximum values of electricity for RO (2–4.5 kWh/m^3) and heat for MD (50–200 kWh/m^3). The electricity source for RO is the Spanish grid and the heat source for MD is heat from a natural gas boiler. Legend: RO-BAU: reverse osmosis – business as usual, RO-GOnorm/RO-GObest: reverse osmosis with graphene oxide-enhanced membranes (normal and best), MD-BAU: membrane distillation – business as usual, MD-GOnorm/MD-GObest: membrane distillation with graphene oxide enhanced membranes (normal and best). Impacts: CCP: climate change potential. FDP: fossil fuel depletion potential. MDP: metal depletion potential. HTPc: human toxicity, cancer. IRP: ionising radiation. EOFP: photochemical ozone formation, ecosystem. HOFP: photochemical ozone formation, human health. ODP: stratospheric ozone depletion. CFC: chlorofluorocarbon. TAP: terrestrial acidification PMFP: particulate matter formation. WDP: water depletion potential. FETP: freshwater ecotoxicity. METP: marine ecotoxicity. TETP: terrestrial ecotoxicity. FEP: freshwater eutrophication. MEP: marine eutrophication. HTPnc: human toxicity, non-cancer. LOP: land use.]

bq. C-60 eq./m^3), which is related to the use of nuclear power in the Spanish electricity mix. The impact reduces with the addition of GO membranes owing to the reduction in the electricity requirements for both RO and MD. The best option is MD-GObest which has a 79 % lower impact that its RO equivalent.

3.1.5. Photochemical oxidants formation potential – ecosystem (EOFP) and human health (HOFP)

The RO-BAU impacts are more than ten times lower than MD-BAU in both categories. This is due to the reliance of MD on natural gas and emissions of nitrous oxides during its combustion. MD-GOnorm and MD-

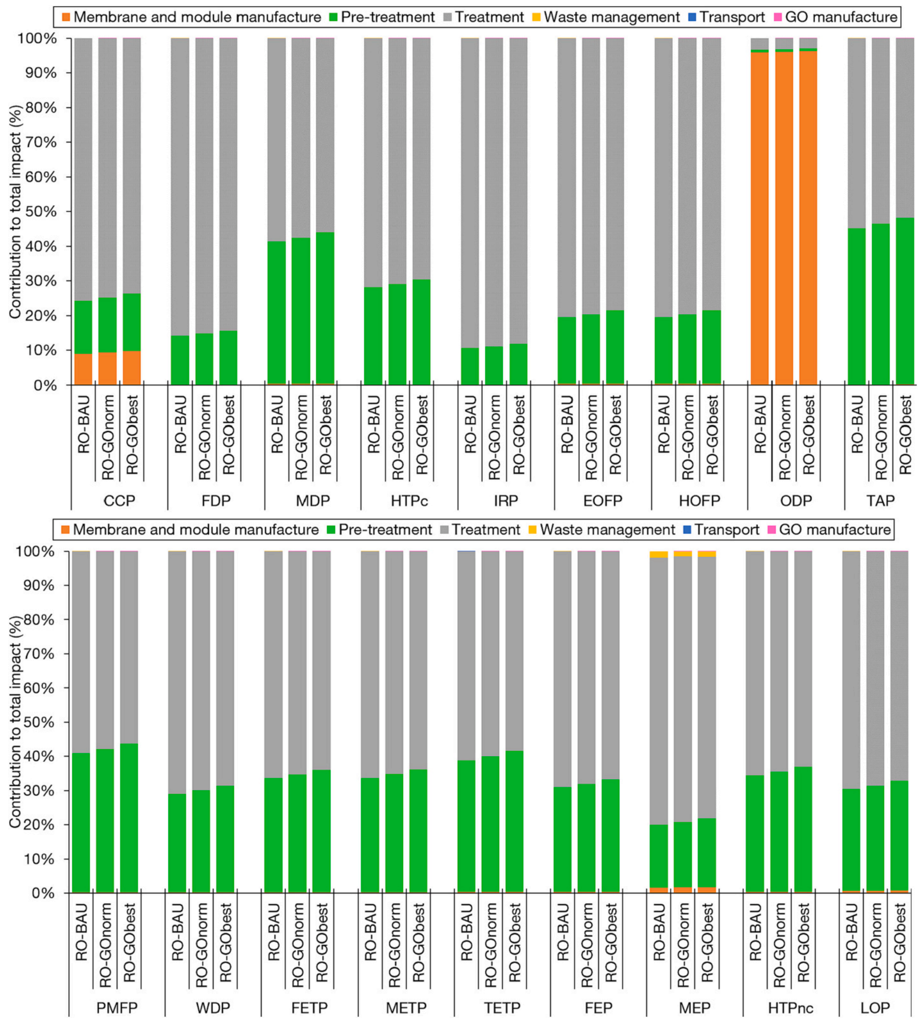

Fig. 5. Contribution analysis for the reverse osmosis scenarios [See caption to Fig. 4 for the abbreviations.].

GObest show a 32 % and 40 % reduction in these impacts relative to MD-BAU. However, they are still significantly above even the RO maximum range.

3.1.6. *Ozone depletion potential (ODP)*

In contrast to most other impacts, MD-BAU has lower ODP than RO-BAU (6.3 compared with 8.8 mg CFC-11 eq./m^3). The difference for the GO-enhanced options is even greater: a 51 % lower impact for MD-GOnorm and 57 % for MD-GObest, relative to their RO counterparts. For MD, the ODP is almost entirely due to the energy consumption in the treatment stage. This is related to HCFs used in some natural-gas production facilities. The impact of RO is mainly attributed to the production of the RO modules, likely due to the release of CFCs, HCFCs and HFCs during module synthesis.

3.1.7. *Terrestrial acidification (TAP)*

TAP of RO-BAU is significantly lower than of MD-BAU (3.2 vs 14 g SO_2 eq./m^3, respectively). This is again due to the use of natural gas in MD and related emissions of nitrous oxides during its combustion, which contribute 95 % to the total impact. For RO-BAU, 45 % is attributed to the pre-treatment stage, most notably to the production of sodium tripolyphosphate, sodium sulphite and sulphuric acid. Adding GO membranes reduces the impact in RO by 2.5–4.3 %. The reduction is much higher in MD, reaching 69 % for MD-GObest. Nevertheless, this is insufficient to bring the MD impact to below that of RO. In both GO-norm and GO-best scenarios, the contribution from the GO manufacturing stage is very small (<0.5 %).

3.1.8. *Particulate matter formation (PMFP)*

This impact is about four times lower for RO-BAU than for MD-BAU (1.2 compared with 4.4 g $PM_{2.5}$ eq./m^3), with the ranges overlapping slightly. For RO-BAU, the largest contribution comes from the electricity consumption (64 %), with the majority of the particulate matter being attributed to the use of hard coal for electricity generation (34 %). The pre-treatment stage accounts for 41 % of the impact, which is mainly associated with the production of sulphuric acid (13 %) and sodium sulphite (14 %). For MD-BAU, the highest contributor is the heat from natural gas (99.6 %).

The use of GO-enhanced membranes could reduce the mean PMFP by 32–40 %, to 3 and 2.7 g $PM_{2.5}$ eq. for MD-GOnorm and MD-GObest, respectively. This overlaps with RO, with the lower range for MD-GObest (1.1 g $PM_{2.5}$ eq.) being below the mean value for RO-BAU and RO-GOnorm (1.18 and 1.14 g $PM_{2.5}$ eq., respectively) and equal to RO-GObest. However, the lower range for MD-GObest is still higher than all the RO lower ranges.

3.1.9. *Water depletion potential (WDP)*

Water depletion is especially important for this study, as it is likely that a seawater desalination plant will be based in a water-scarce region. This impact is 31.6 % higher for MD-BAU than the equivalent RO option (1.9 and 1.3 L/m^3, respectively). For the latter, the largest contributor is electricity (74 %) owing to the use of hydropower (32 %) and nuclear

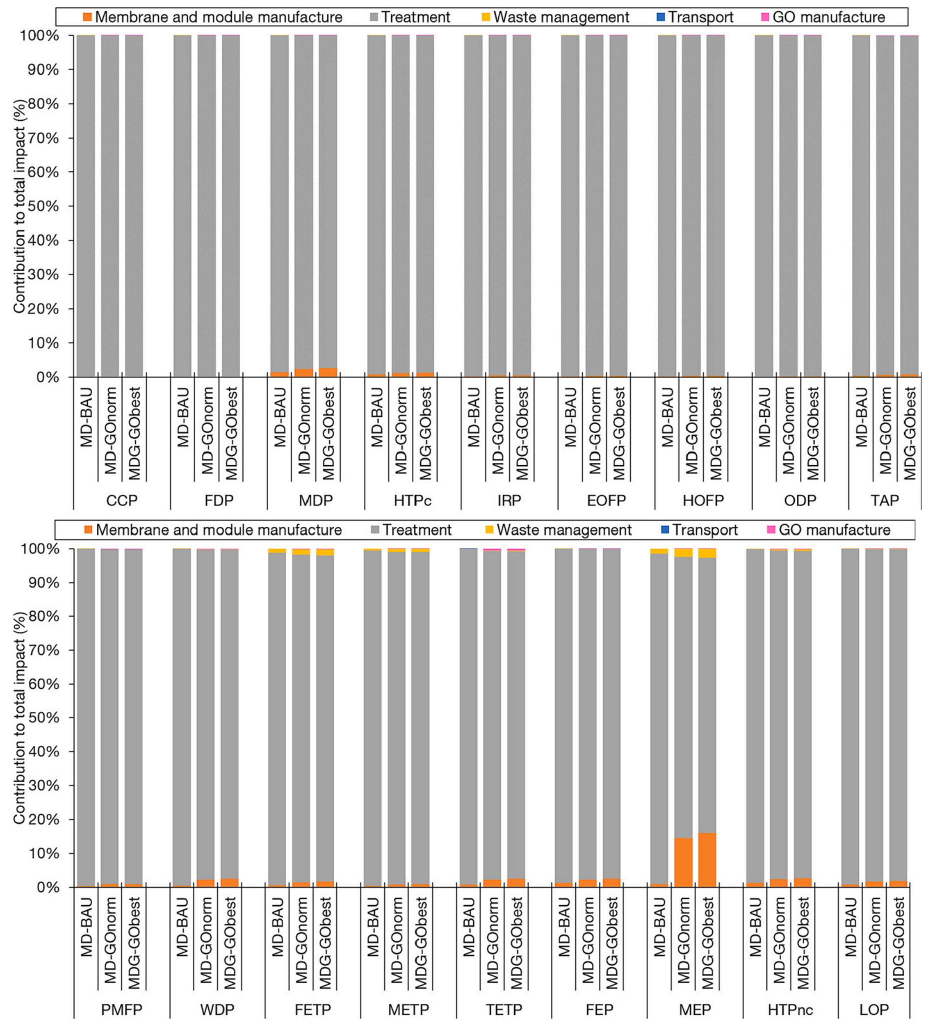

Fig. 6. Contribution analysis for the membrane distillation scenarios [See caption to Fig. 4 for the abbreviations.].

energy (19 %) in the grid. For MD-BAU, the heat accounts for 80 % of WDP.

The addition of GO-membranes reduces water depletion in both RO and MD, with greater reductions in the latter. This is due to the reduction in the energy consumption for water treatment, which results in MD-GObest having the lowest WDP among all the options (1.2 L/m^3). This shows that it is possible to produce 1000 L of drinking water while consuming just over 1 L of freshwater.

3.1.10. Ecotoxicity potential – freshwater (FETP), marine (METP) and terrestrial (TETP)

The freshwater ecotoxicity is two times lower for RO-BAU (0.18 g 1.4-DB eq./m^3) than for its MD equivalent (0.36 g 1.4-DB eq./m^3). The consumption of electricity for water treatment accounts for 68 % of the impact, of which 46 % is due to the production of wind turbines. The pre-treatment stage accounts for 32 % of the total FETP and the largest contributors are the electricity consumption, the production of sodium tripolyphosphate and sulphuric acid. For MD-BAU, the largest contribution comes from the extraction of natural gas (82 %), followed by the manufacture of sodium tripolyphosphate (13 %). Similar trends can be seen for marine ecotoxicity, with 0.23 g and 0.85 g 1.4-DB eq./m^3 for RO-BAU and MD-BAU, respectively. GO-enhanced membranes reduce both impacts in RO and MD. However, despite the reduction of 39 % in MD-GObest, its impacts are still higher than those of any other RO options.

For terrestrial ecotoxicity, the results are similar for RO-BAU and MD-BAU, at 1.36 and 1.38 kg 1.4-DB eq./m^3, respectively. In terms of contribution, 20 % of RO's terrestrial ecotoxicity can be attributed to the use of sulphuric acid for pH adjustment, as RO membranes need additional pre-treatment [31]. For the RO-GOnorm and RO-GObest scenarios, the TETP is 1.32 and 1.27 kg 1.4-DB eq./m^3, respectively, which is lower than for MD-BAU owing to the reduction in the required electrical energy. However, MD-GOnorm and MD-GObest have lower TETP than their RO equivalents (1.1 and 0.99 kg 1.4-DB eq./m^3). This is because GO membranes enable a greater reduction in the energy consumption. MD-GObest at the lower range achieves the lowest impacts for both terrestrial and freshwater ecotoxicity (0.38 and 0.88 g 1.4-DB eq./m^3). This implies that newer, more efficient MD plants that use GO-enhanced membranes can enable desalination at lower impacts in these categories than RO-GO installations.

3.1.11. Eutrophication potential – freshwater (FEP) and marine (MEP)

At 0.17 g P eq./m^3, the FEP of RO-BAU is lower by a factor of two than that of MD-BAU (0.33 g P eq./m^3). The MEP of RO-BAU is also lower than that of its MD equivalent (0.028 vs 0.034 g N eq./m^3). Similar to the other impacts, the main contributors to both impacts are electricity used in RO and heat used in MD.

The lowest MEP is found for MD-GObest and RO-GObest (26 mg N eq./m^3). The use of GO-enhanced membranes has a more significant effect for freshwater eutrophication and, as a result, at the lower range, the FEP of MD-GOnorm (88 mg P eq./m^3) and MD-GObest (79 mg P eq./m^3) is lower than that of any of the RO scenarios.

3.1.12. Land use potential (LOP)

Less land is required for RO-BAU (0.016 annual crop eq.·yr) than for MD-BAU (0.025 annual crop eq.·yr). The use of land is related to the life cycle of energy used for water treatment, which accounts for 72 % and 83 % of the impact for RO-BAU and MD-BAU, respectively. The largest single contributor for RO-BAU is the electricity generation via solar PV, as these are ground installations. For MD-BAU, the impact is largely due to the disruption of land during extraction of natural gas. The lowest impact is achieved with MD-GObest (0.005 annual crop eq.·yr) at the lower range, half the lower-range value for the best RO-GO option (0.011 annual crop eq.·yr).

3.2. Sensitivity analysis

This section explores the effect on the results of the electricity mix used for RO and of heat sources in MD as energy consumption in both is a significant contributor to the impacts.

3.2.1. Electricity sources for RO

The electricity mix has a significant effect on the RO impacts as shown in Fig. 7. As expected, grid mixes that rely on non-renewable energy sources (e.g. United Arab Emirates (UAE) and Saudi Arabia) have much higher impacts than those with greater contribution of renewables (e.g. Spain and California). Among the electricity options considered, using the Spanish grid leads to the lowest climate change (0.97 kg CO_2 eq./m^3) and ozone depletion (8.8 mg CFC-11 eq./m^3) potentials. However, situating the RO plant in the UAE would results in the lowest impacts for 12 categories; this is due to the high contribution of natural gas in the national grid (Table 3) which has lower burdens for those impacts than the renewables present in the other electricity mixes considered here. California is the best option for three impacts (fossil fuel depletion, ecosystem and human health photochemical oxidant formation) and Saudi Arabia for one (ionising radiation). The climate change potential for the Saudi electricity mix is over four times that of the Spanish grid (3.9 and 0.97 kg CO_2 eq./m^3, respectively); it also has the highest impacts for nine categories. This is due to over 99 % of its electricity being sourced from oil and natural gas, while the Spanish grid has a higher percentage of renewables, such as wind (21 %) and solar (8 %). The Californian electricity shows some of the highest impacts for freshwater (0.64 g P eq./m^3) and marine eutrophication (0.087 g N eq./m^3), human toxicity - cancer (7.3 g 1,4-DB eq./m^3) and land use (0.056 annual crop eq.·yr). This is mainly due to the imported electricity from the rest of the US, which is reliant on coal (23 %). The high land use is related to the generation of heat and power from wood chips.

3.2.2. Heat sources for MD

Four different thermal energy sources were evaluated: natural gas boiler, solar thermal, biogas and waste heat. Across all 18 impact categories, the waste heat emerged as the best option (Fig. 8). This is because the impacts from the treatment stage are reduced sharply as all the thermal energy requirements are met using waste heat. However, this system is limited by location and consistent heat supply. If waste heat is unavailable, solar thermal should be considered as an alternative as it has the lowest impacts for ten categories, including the climate change potential (1.4 kg CO_2 eq./m^3). However, it also has highest freshwater ecotoxicity and metal depletion related to the metals used to manufacture the solar panels. Using biogas gives mixed results: it has lower impacts than natural gas for seven categories (including CCP of 5.5 kg CO_2 eq./m^3) but is the worst option for seven categories, including particulate matter formation and freshwater eutrophication.

Comparing the results for RO and MD shows that an MD desalination plant using solar-thermal energy would have 43–93 % lower impacts for nine categories, including a 64 % lower CCP, than an RO plant powered by the Saudi grid. These findings indicate that, if the grid mix in the location of the proposed desalination plant consists predominately of fossil fuels, such as Saudi Arabia which relies globally the most on seawater desalination, then MD using solar thermal would be the preferred choice. Other options include generating the required

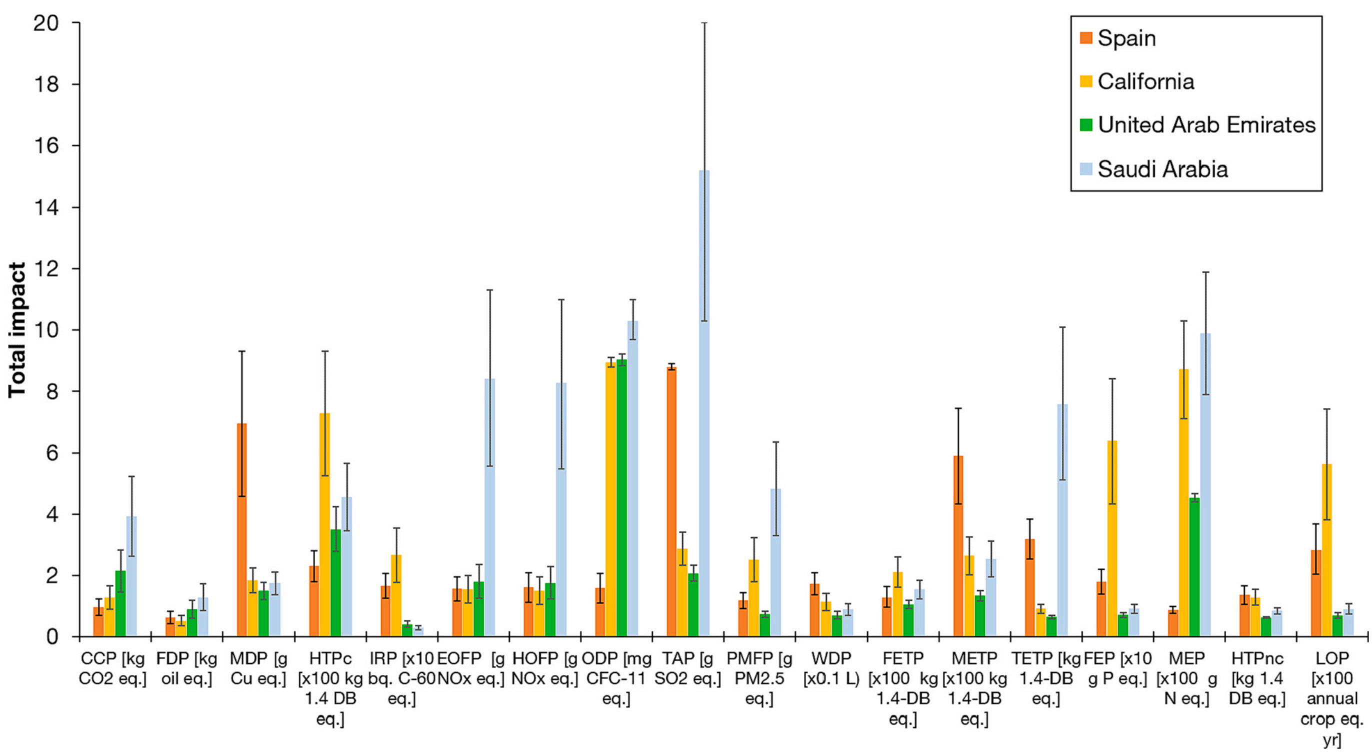

Fig. 7. Environmental impacts of reverse osmosis for selected electrical sources of electricity [The impacts are expressed per 1 m^3 of potable water. The error bars represent the minimum and maximum values of the consumption of electricity for RO (2–4.5 kWh/m^3) and heat for MD (50–200 kWh/m^3). See caption to Fig. 4 for the abbreviations.].

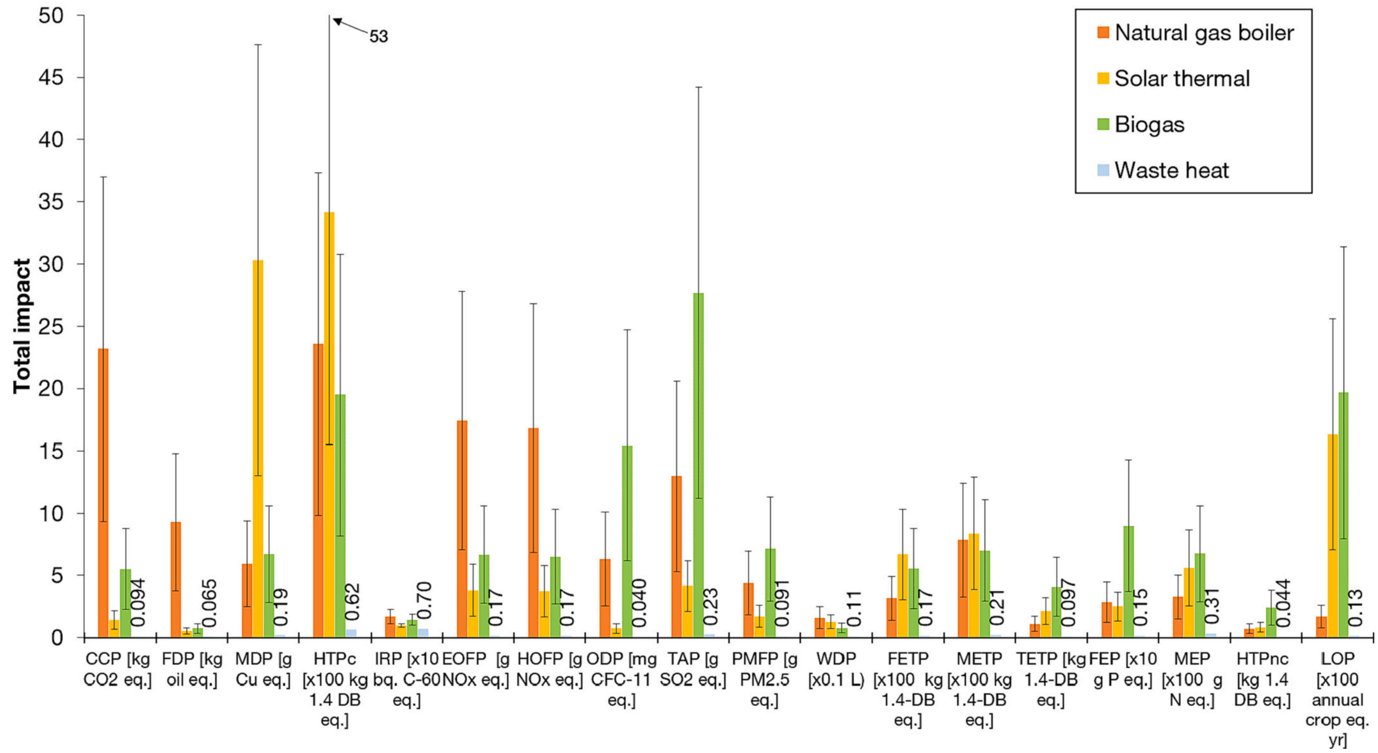

Fig. 8. Environmental impacts of membrane distillation for selected heat sources [The impacts are expressed per 1 m^3 of potable water. The error bars represent the minimum and maximum values of the consumption of electricity for RO (2–4.5 kWh/m^3) and heat for MD (50–200 kWh/m^3). See caption to Fig. 4 for the abbreviations.].

electricity from an off-grid supply of renewable energy, which is currently being explored in the Arabian Gulf.

3.3. Comparison of results with literature

A comparison with other LCA studies of RO and MD that use GO-enhanced membranes for seawater desalination is not possible as such studies are not available. The only two other GO LCA studies are for vastly different systems (filtration of palm oil mill effluent [64] and an algal membrane photoreactor [50]). Nevertheless, they also report that the process improvements offset the GO manufacturing impacts, congruent with the findings of the current study.

Direct comparisons of results with other works for RO and MD without GO are also difficult owing to differences in the goal and scope, impact assessment methods, scale of the plants and geographical locations. However, an attempt at comparison with literature was made as discussed below.

The CCP range for RO found in this study (0.69–5.2 kg CO_2 eq./m^3) is in a relatively good agreement with the values reported in the literature (1.8–6.1 kg CO_2 eq./m^3) [13,15,69]. However, looking at the comparisons of individual studies in Fig. 9a, it can be seen that the RO-BAU impact in this study is nearly twice that reported by Tarnacki et al. [15], despite both being based in Spain. Given that the Tarnacki study is more than a decade old, this could be due to the transition to renewable energy in Spain since then.

The results in the current study show that the location of the RO facility affects the CCP significantly due to the local energy mix. This corresponds to the findings by Al-Kaabi et al. [13], who evaluated solar-

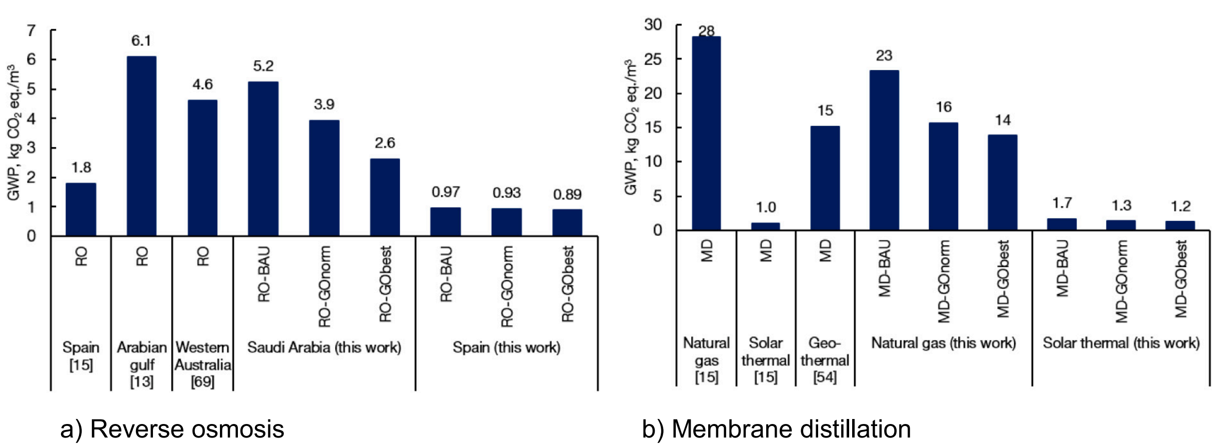

Fig. 9. Comparison of the global warming potential (GWP) estimated in the literature and the current study for reverse osmosis (a) and membrane distillation (b) [The "GWP" in the current study is referred to as the "CCP" (climate change potential), in congruence with the ReCiPe method.].

PV powered RO against natural gas and heavy fuel oil in the Arabian Gulf. The CCP of the solar-PV scenario was >90 % lower than of fossil-fuel options; however, the ODP and ADP were significantly higher, which is in agreement with the results in the current work.

As seen in Fig. 9b, the CCP range for the MD scenarios obtained in the current work (0.17–23 kg CO_2 eq./m^3) is also mostly within the literature range (1–28 kg CO_2 eq./m^3). Comparing the results for natural gas, Tarnacki et al. [15] reported a CCP of 28 kg CO_2 eq./m^3, relatively close to 23 kg CO_2 eq./m^3 estimated here. This is despite the energy consumption in Tarnacki et al. being less than half (56 kWh/m^3) that in this study (125 kWh/m^3). This is reflected in their results for solar thermal, which are 70 % lower than those found in the current work (1 vs 1.7 kWh/m^3). Furthermore, the impacts of MD-GO using natural gas are comparable with the study by Liang et al. [54] who assumed the use of geothermal energy in MD (without GO), but it is unclear what energy consumption was assumed in their model. However, in contrast with the findings here and in Tarnacki et al. [15], Siefan et al. [66] found that solar PV had higher CCP (for some PV types) than the system relying on grid power (not shown in the figure as the authors only reported percentage rather than absolute values). Nonetheless, the rest of the 20 impacts considered by Siefan et al. were lower for the solar-PV scenario.

3.4. Study limitations and future research

The energy consumed during water abstraction and distribution was outside of the scope in this study as it would be the same for both RO and MD. Additionally, the impacts of brine disposal were not evaluated, but recent studies suggest that high salinity brine can have damaging effects on local marine biodiversity [122]. The lower water recovery from the incoming seawater in MD (5 %) compared with RO (40 %) may address this issue, as the brine has a lower concentration of total dissolved solids [29,123]. However, the lower recovery has a penalty: the seawater throughput is higher which requires a larger amount of anti-scalant. The latter (sodium tripolyphosphate) contributes significantly to the freshwater, marine and terrestrial eutrophication and toxicity impacts. This aspect needs to be investigated further, particularly as some studies suggest that MD can use lower quantities of the anti-scalant [31,124].

The impacts of the brine disposal could also be reduced by incorporating a hybrid RO-MD system whereby the RO brine is used as the MD feed. This would also recover additional potable water, but there is still the issue of waste brine and the cumulative energy demand for both systems can be 22–67 kWh/m^3 potable water [125]. Membrane crystallisers could be used in series with an RO-MD hybrid unit to recover completely all of the water and produce salt [18]. As the membrane crystallisers require thermal energy for the additional heater, as well as mechanical energy to drive the Carnot cycle [126], they should only be considered in locations near to sensitive sea areas to avoid the energy penalty.

In relation to technical aspects of membranes for MD, future research should investigate how GO can be applied to enhance further the permeate flux, as this reduces the thermal energy requirements. The anti-scaling and 'self-cleaning' properties of GO membranes should also be explored to determine whether implementing these membranes could reduce consumption of anti-scalant, or enable the use of less aggressive chemicals [127,128]. This would be beneficial particularly for RO, where the pre-treatment and cleaning stages account for 20 % of the impact in 11 categories.

This study is also limited by current knowledge of the health and environmental impacts of nanomaterials [129]. Although it is expected that GO would be stable and would not leach when disposed in landfill, the decomposition behaviour of GO is not well understood, which could affect the reliability of the results in this study related to waste management. In addition, the synthesis route to produce GO-enhanced membranes is not well defined. A range of different solvents, dispersion agents and binders (which were excluded as LCA data were not available) are used in literature [50,130,131] and this could have an effect on the LCA results. Synthesis of GO from waste is also being explored [132] and future research should investigate how this may affect the LCA results.

Furthermore, the lack of ecotoxicity data for GO hinders the ability to assess these impacts. Deng et al. [133] derived them for freshwater ecotoxicity of GO but found that the impact ranged by three orders of magnitude ($1–10^3$ potentially affected species day m^3 kg^{-1}). By comparing the results with other nanomaterials, the authors also found that GO had a significantly lower toxicity than carbon nanotubes and nano-silver.

It is also worth mentioning that the energy consumption of RO is not expected to decrease significantly below the minimum range considered in this study (2 kWh/m^3). Conversely, improvements in process design are leading to step-wise reductions in the energy consumption for MD and hence future studies should consider the related effect on the environmental impacts of MD.

4. Conclusions

This paper presented an LCA of seawater desalination by RO and MD, considering both techniques with and without GO-enhanced membranes. In comparison to MD, RO was overall the best option with or without the GO membranes. The addition of GO led to better performance in both technologies due to its anti-fouling (RO) and high-permeability (MD) properties, hence reducing the impacts. For MD, the gains of implementing GO membranes were much greater than in RO, lowering the impacts on average by 27–34 % (vs 3–6.8 % in RO). Nevertheless, even these small improvements in RO could still lead to a significant reduction in GHG emissions if GO membranes were implemented across all existing seawater RO plants (380,000–850,000 t CO_2 eq. per year). The main contributor to the impacts in both technologies (>90 %) is energy consumption. The contribution of the production of GO membranes is very small (0.02–0.59 %).

The sensitivity analysis revealed that situating RO plants in regions with higher contribution of renewables in the grid would lead to the lowest climate change and fossil depletion impacts. However, operating them in regions dominated by natural-gas electricity would be a better option for most other impacts. For MD, the lowest impacts would be achieved if using waste heat. Using solar-thermal energy instead of natural gas would lead to 43–93 % lower impacts of MD than RO powered predominantly by fossil fuels, including climate change, which would be 64 % lower.

These results suggest that future research should focus on developing GO membranes that reduce the energy required for the treatment stage. For RO (which is already at the lower flux limit), this is through the development of anti-fouling and self-cleaning membranes. For MD, this is through the development of high-permeability membranes.

Furthermore, the main concerns that need to be addressed before potential commercialisation of GO-enhanced membranes for RO and MD include proof of immobilisation on support membranes, ensuring leaching of nanoparticles into drinking water does not occur, and the long-term stability testing of GO membranes for desalination (including adequate cleaning regimes). Future LCA studies should consider different GO membrane synthesis methods (such as electrospinning), as well as alternative ways that GO can be added to membranes to enhance their performance (for example, laminate membranes). Other desalination systems could also be considered, for example a hybrid RO-MD system.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.desal.2023.116418.

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