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Biogas from source separated organic waste within a circular and life cycle perspective. A case study in Ontario, Canada



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ABSTRACT

The appropriate transformation and valorisation of biogas offers environmental and economic opportunities in a future with restrictions upon fossil-based fuels and materials. The LCA method was used to quantify and compare the potential environmental impacts of an AD plant incorporating biogas co-generation and upgrading options, namely AD-CHP and AD-RNG. Using an average Anaerobic Digestion facility in Ontario, Canada, modelled after real facilities, as a case study, electricity and steel were identified as potential hotspot input materials carrying a disproportionate environmental burden for biogas production. With a system expansion approach, the biogas was subsequently utilized to produce (1) both heat and electricity using a Combined Heat and Power (CHP) system, or (2) upgraded to renewable natural gas (also called biomethane) through chemical amine scrubbing, respectively.

In comparing the biogas co-generation and upgrading options, the AD-CHP alternative resulted in a lesser environmental load, two times lower when compared to the AD-RNG biomethane recovery option. Furthermore, the avoided burden of producing fossil-based electricity, natural gas, and chemical fertilizer was analyzed and compared against their renewable counterparts. Significant reductions in emissions and in the depletion of fossil fuels were achieved, thus confirming the positive efforts of diverting organic waste from landfills to reduce organic waste disposal impacts and improve the management of organic waste. The analysis has provided useful insights to bioenergy project developers, policy makers and the scientific community regarding the processing of source separated organic waste, biogas production, and its upgrading alternatives in a circular economy perspective.

1. Introduction

A worldwide population of more than 7.8 billion people (United Nations, Department of Economic and Social Affairs, 2019), coupled with rapid developments in some regions and improved freedoms/welfare, has led to increased consumption of resources and the generation of large quantities of waste (Cerda et al., 2018). If the current global trend continues, aided by rapid urbanization, billions of metric tonnes of municipal waste management authorities (Sharholy et al., 2008, Zhou et al., 2018). Currently, approximately over 2.01 billion tonnes of municipal wastes are generated worldwide and by 2050, about 3.4

billion tonnes of municipal waste is anticipated (Paes et al., 2019). According to the internationally-recognised Food and Agriculture Organisation (FAO), more than a third of food that is produced and distributed from "farm to fork" is lost (FAO, 2018). Particularly in the developed world where most food losses occur during processing, storage, and consumption, while in developing countries, losses primarily occurring during the production phase (European Commission, 2018; FAO, 2018). Generated waste streams vary in specific parameters such as physical state, odor, toxicity, and organic content (Arafat et al., 2015).

In the Province of Ontario, Canada, 4 million metric tonnes of organic waste are generated annually out of the total 12 million metric

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Received 1 February 2021; Received in revised form 16 June 2021; Accepted 6 July 2021 Available online 9 July 2021 2665-9727/© 2021 The Authors. Published by Elsevier Inc. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-ad/4.0/). tonnes of garbage, which, when combined with the rapidly diminishing landfill capacity, represents a challenge to the economy, environment, and society (OWMA, 2015; FAO, 2018). At present, due to insufficient infrastructure and regulatory impetus, much of the organic waste eventually ends up at local landfills and some is trucked to the United States of America (USA) for final disposal. In addition, budgetary restrictions for infrastructure developments in waste management have led to improper treatment of waste, and the most commonly applied technologies are still linear, such as landfilling and incineration (Wainaina et al., 2020). A shift towards the development of renewable energy sources supported by materializing price drivers has increased in recent years, due to the depletion of, and environmental concerns over, the continued reliance upon conventional fossil fuels such as coal and petroleum (Cherubini and Ulgiati, 2010; Christian, 2000). Organic waste is expected to play a pivotal role as a valuable feedstock material when converted into a cost-effective renewable energy (CBA, 2015; Ma and Liu, 2019). When the organic waste material is diverted from landfills, it brings environmental benefits (Environment Canada, 2013). Many nations and local authorities likewise, are embracing the concept of a Circular Economy (CE) by attempting to divert waste from landfills and, at the same time, reducing the environmental impact of consumption by bringing into practice advanced and sustainable efficient ways in managing the end-of-life of materials (Yadav and Samadder, 2018; González-García et al., 2019).

In 2016, Canada became the first country in the Americas to enact a comprehensive circular economy law, consisting of legislation that will tackle the problem of waste by increasing resource recovery and the elimination of waste (Ghosh, 2019). Although the concept of transforming what would normally be disposed as waste into useable energy, chemicals, and fuels is not new, its applicability has expanded considerably in recent years even in waste treatment facilities. Within a CE narrative, the value of products, materials, and resources is kept in the economy for as long as possible, generating new materials, while minimising waste (European Commission, 2015; Ellen MacArthur Foundation, 2012). Acting in parallel, a new model called the circular bioeconomy considers the potential of bioresources to generate renewable biological resources and their conversion into value-added bio-based products (Mougenot and Doussoulin, 2021). Within this bio-based model, biomass waste such as source separated organics (SSO) can represent an opportunity to recover valuable nutrients to enrich and improve soil quality using compost or digestate, and/or to recover bioenergy and bio-based products (OWMA, 2015). The circular bioeconomy concept is therefore, expected to contribute towards sustainability and to complement and act in parallel towards supporting the aspirations and principles of CE (Ubando et al., 2020). However, declaring such developments towards less carbon intense transitions as environmentally preferable should be approached with caution, as there is yet no harmonised method to assess whether a specific strategy truly contributes towards sustainability or not (Zucaro et al., 2019; Blum et al., 2020).

1.1. The rise of commercial scale anaerobic digestion plants in Ontario

Among several renewable energies, biogas is projected as the most formidable renewable energy source (Singh et al., 2020). Anaerobic digestion (AD) facilities are expected to increase the value of SSO, predominantly from agro-industrial, residential, and industrial sources to produce biogas (Ericsson et al., 2020). AD technology in enclosed bioreactors provides new opportunities to capture the much-needed renewable energy from the decomposition of organic materials. AD is the enclosed, oxygen-free biodegradation process that generates the methane-rich biogas, that can be used to generate either renewable natural gas (RNG) or combusted to produce electricity and/or heat (CHP) (Bátori et al., 2018). For example, a biogas plant with an electrical power output of 2.5 MW, has the potential to provide enough energy to power about 2500 homes, with an expected reduction in greenhouse gas emissions of more than 224,000 metric tonnes CO_2 equivalent by 2020 (Kryzanowski, 2020). The capture of energy from waste diverted from landfills also reduces odor and this renewable energy solution could produce about 3% of the natural gas demand in Canada or the equivalent of 1.3% (equal to 820 MW) of the electrical power needs using reliable, clean, green electricity obtained without the use of energy crops or fossil fuels (Kelleher Robins, 2013). According to a Canadian biogas study technical report in 2013, biogas production processes have the potential to reduce Canada's GHG emissions by 37.5 million tonnes CO_2 eq per year, which is equivalent to the removal of 7.5 million cars off the road (Canadian Biogas Association, 2015; Kelleher Robins, 2013).

Organic waste diversion from landfills also provides financial benefits (Ontario Federation of Agriculture, 2015, White et al., 2011). The major economic benefit came through in 2007, because of the Ontario Feed-In Tariff (FIT) program which subsequently led to the growth and deployment of several commercial-scale AD projects in Ontario. Moreover, the opportunities for selling the nutrient-rich digestate residue as a bio-fertilizer was considered economically beneficial in addition to returning nutrients to the soil (Albanna, 2013). The proper processing of SSO waste through the production of biofertilisers enhances soil fertility, thus supporting the sustainability of Ontario's agricultural sector (Ontario Federation of Agriculture, 2015). Additionally, due to changes in federal policies, there has been increased focus on the substitution of natural gas with renewable natural gas produced from biogas using upgrading technologies. In a 2014 report, the Conference Board of Canada provided an estimated increase in job creation (13,000 net new jobs) in Ontario and GDP increase by \$1.5 billion through waste diversion (CBC, 2014). However, in contrast to the dynamic growth projections for AD, there has been modest corresponding developments of environmental standards, particularly for the invasive materials, such as commercial food packaging materials. The presence of contaminants in the feedstock, such as plastic bags, is a major technical challenge for AD facilities (Bátori et al., 2018). The residual material's feasibility for composting and field application can also be a concern due to the presence of heavy metals, organic pollutants, and emerging contaminants that may accumulate in soils (Yang et al., 2017). According to Awasthi et al. (2018), higher levels of heavy metals in organic residues from sewage treatment have been recorded. Although not commonly used as feedstocks for food or organic waste digesters, sewage residues are anaerobically digested in some areas prior to agricultural application. There is also a growing interest in co-digesting food waste at municipal sewage treatment plants, although this is not currently being done on a large scale. Therefore, strict regulations and standards are required to regulate the maximum permissible values in material applied onto agricultural lands to ensure food security (Wainaina et al., 2020).

All CE supporting activities such as the production of biogas should be assessed in the light of economic, environmental, and social sustainability. A good understanding of the different needs and values of the affected stakeholders is necessary when selecting tools for sustainability assessments (Gasparatos et al., 2012; Blum et al., 2020; Peña et al., 2020). In fact, the chosen mechanisms and methodologies (or "tools") frame the sustainability assessment and carry ethical and practical implications (Gasparatos et al., 2012). However, despite the ongoing concerted academic effort in CE, disagreements still exists on how to measure the transition towards sustainability and circular economy (Gasparatos et al., 2012). In this study, Life Cycle Assessment (LCA) has been chosen as the most appropriate tool. LCA has a non-reductionist approach with more than eighteen impact categories and applies a consumer-side perspective, capable of including all the process phases in terms of their direct impacts on the investigated areas (Santagata et al., 2020a,b). According to Christensen et al. (2020), LCA is expected to play a critical role towards integrated waste management to understanding and improving existing waste processing facilities, comparing alternative technologies, and understanding their performance for strategic policy development. Consequently, a look into

policy provides an opportunity for coherence in regulation development that supports waste management, AD operations, and transitions towards sustainable circular economy by providing added value for practical implementation, as well as addressing the needs of the affected stakeholders through organic waste reduction, processing, and recovery of renewable energy and bio-based products.

At the intersection of academia and practice, the CE concept is based on the notion that, the more a material is kept in circulation, the better. However, this may not necessarily equate to greater sustainability, according to Blum et al. (2020). Therefore, insights from different academic disciplines, in this case, environmental performance evaluated through LCA to generate policy-oriented perspectives, are relevant when developing solutions for sustainable waste management and processing in AD facilities. Much of the scientific literature has examined the environmental performance of AD facilities with particular focus on China and Europe (Linville et al., 2015; Horváth et al., 2016; Bacenetti et al., 2016). This body of literature has over the years, provided decision makers with solid background knowledge on how to improve efficiency of AD systems and waste management (Vosooghnia et al., 2021). However, according to Hijazi et al. (2016) due to dissimilar functional units and the type of feedstock biomass materials, direct comparisons of the previous studies are still difficult to perform from an environmental point of view. Although there are several specific LCA focused studies on anaerobic digestion (Blengini et al., 2011; Bühle et al., 2011; Whiting and Azapagic, 2014), to the best of our knowledge, very few studies have considered an entire analysis of biogas primary production from SSO to its upgrade into renewable natural gas and co-generation into heat/or electricity at the same time. Through life cycle impact assessment (LCIA) this study also contributes to providing considerable information on comparative environmental outcomes between biogas co-generation and upgrading alternatives. Moreover, there is a current knowledge gap on site-specific case studies considering the processing of SSO at AD facilities, particularly in North America (Wang et al., 2021). Furthermore, this study can be considered timely due to the emerging and current policy drivers that seem to deliberately favour and support developments in renewable energy worldwide. As the global bioenergy sector advances in response to climate change and the depletion of non-renewable energy sources, this study presents a huge opportunity for the Canadian environmental sector. In addition, environmental performance evaluations capable of comparing existing AD with biogas upgrading technologies are much needed to eliminate irrational policy practices, by providing quantified evidence on environmental performance and circular economy perspectives (Florio et al., 2019). Against this background, the aim of this study is to therefore expand the focus from not only the production of biogas but to include biogas valorisation alternatives towards the recovery of heat and/electricity or biomethane plus biofertiliser. The deployment of an LCA perspective in this study helps to understand, in critical terms, the processing of organic waste and its potential to provide future generations with renewable energy. Furthermore, for increased environmental awareness, this study identifies hotspot areas from the processing of SSO and highlights the additional environmental benefits that can be derived from the substitution of fossil-based natural gas, electricity, heat, and chemical fertilizers.

In summary, this study progresses and contributes to the scientific literature on the management of organic waste and increased circularity approaches in AD facilities. The effort to divert organic waste from landfills towards AD facilities is expected to bring positive environmental benefits. The use of LCA offers an environmental perspective to inform policies around the management and treatment of SSO. This approach is considered relevant to Canada as the available space in existing landfills is becoming increasingly scarce, coupled with the challenges related to public approval, obtaining permits, and siting of new landfills. The landfill capacity deficit in Ontario results in the exportation of significant quantities of waste to the United States of America and this dependency on foreign landfill capacity does not represent a sustainable framework, especially considering the existing uncertainties in the permissible transfer of waste across international borders. Therefore, cooperation between regulators (both provincial and federal) and researchers across Canada and globally is needed in developing coherent circular approaches in pursuing organic waste reduction and the further development of biogas upgrading projects.

2. Materials and methods

2.1. Case study description

The investigated case study explores the production of biogas, electricity, heat, and RNG derived from AD in Ontario, Canada. This case study examines an average and representative medium commercial size (or meso-scale) waste management and recycling services company, based on information collected from real and currently operational facilities.¹ Average data from these real facilities was used to create a more robust closed-loop system in Ontario, Canada. The representative facility operates with commercial scale, completely mixed wet anaerobic digesters to process the mixed organic waste stream. The maximum quantity of solid and liquid waste delivered to the investigated site is 75,000 metric tonnes per year, as set by provincial environmental permitting requirements. Private transporters are responsible for delivering the waste to the processing facilities and the environmental burden of collecting SSO is not considered in this study due to lack of data.

Leaning slightly away from the traditional trend towards on-farm animal manure digestion, the investigated site accepts by-products from fruit and vegetable processing, grains, fish, meat, fats, oils, and grease (FOG), prepared foods manufacturing, and dairy by-products from residential, retail, and industrial areas within an approximately 100 km radius. The arriving wet food waste, other organic waste, and SSO material is processed using separation equipment. The purpose of the separation equipment is to separate inorganic materials, such as plastic bags and retail packaging from organics using mechanical paddles or other methods. This process also removes heavy solids such as bone fragments, metals, glass, and ceramics. The removed material is sent to landfill, while the clean organic slurry feedstock continues through the rest of the process. Biofilters are used to treat the odorous air from the facility before being released into the atmosphere. The organic slurry is pasteurized and stored, before being fed into the anaerobic digester tanks. Feeding the digester tanks is controlled by the process operator to maintain the correct biological balance within the AD system. Three digester tanks are used for the anaerobic digestion process to produce biogas, which is immediately transferred to the on-site CHP facility with an electricity generation capacity of 2.5 MW. In addition to electricity, heat energy is captured for pasteurizing the raw organic slurry, warming the digesters, and feedstock. Other on-site uses of excess heat include preheating digestate material to reduce volume, ultimately saving on storage and transportation costs, while retaining all the nutritional benefits contained in the digestate. Any remaining pathogens are destroyed during the drying process and the remaining material is then pelletized to make biofertiliser. The organic fertilizer has 80% organic content and is rich in nitrogen, phosphorus, and potassium (NPK).

2.2. Goal and scope definition of the LCA study

To carry out a comprehensive assessment of the environmental impact associated with a product, process, or service, the standardized LCA methodology is preferred (Fiorentino et al., 2015). A cradle-to-gate approach is applied since the system boundary in this study is restricted

¹ To protect proprietary and confidential information, including the specific inventory data discussed in this case study, we are not able to disclose the identity of the facilities.

and limited to the production of biogas and its conversion into electricity and heat (AD-CHP), while neglecting the use stage (Florio et al., 2019; Pellegrini et al., 2015). Due to the increased focus on RNG in Canada, the average representative site was further expanded, to include the upgrade of biogas to renewable natural gas (AD-RNG) in order to provide useful insights to developers and policy makers on accelerating RNG project development (Canadian Biogas Association, 2021). The boundary of the examined system is shown in Fig. 1.

The following interrelated components of the LCA study are organized according to the ISO standards (2006 a, b): (i) goal and scope definition; (ii) inventory analysis; (iii) impact assessment, and (iv) interpretation of results for explanation of conclusions and recommendations. The goal of this study is to estimate the environmental burdens generated from the processing of organic waste at a modelled average representative facility and to compare, from an environmental perspective, the different biogas co-generation and upgrade options (AD-CHP and AD-RNG). The avoided burden of producing fossil-based electricity, heat, renewable natural gas, and chemical fertilizer is also included in the analysis.

The AD plant processes approximately 205 metric tonnes of SSO feedstock material that is delivered to the site per day. The functional unit for the study is based on the main energy product produced at the site, namely MWhel (electricity) production. This electricity, measured as MWhel, production is produced by the onsite combined heat and power (CHP) co-generation system. The CHP consists of an internal combustion biogas-powered engine that drives an electrical generator, and the produced electrical power is then sold to the local electrical utility company. Therefore, all supporting infrastructure and equipment such as concrete, steel and asphalt were calculated based on MWhel. In order to compare the AD-CHP co-generation system and the AD-RNG upgrade systems, the processing of 1 metric tonne of wet SSO which produces 87.43 m³ of CH₄ plus 48.96 m³ of CO₂ was considered (205 tonne wet SSO processed per day generates 61.60 MWhel production per day and this translates into 0.3 MWhel per wet tonne of SSO). Several referenced studies have employed an input based approach using one wet tonne of organic waste processed to compare different alternatives (Nordahl et al., 2020; Opatokun et al., 2017). According to the International Reference Life Cycle Data System (ILCD) Handbook (International Reference Life Cycle Data System Handbook, 2010), the analyzed context can take into account the environmental impacts of the entity

which is running the biogas plant for micro-level decision making and as such an attributional Life Cycle Inventory (LCI) modeling framework is adopted (Fiorentino et al., 2015; Bartocci et al., 2020).

2.3. LCA inventory acquisition and impact analysis

The inventory data for the AD-CHP process were obtained through site visits and physical interactions with cooperating organic waste processing operators based in Ontario. The obtained primary data was then averaged to create a representative reference site, referred to in this study. One of the visited sites is a public facility, operated by the municipality of Toronto, to process organic waste. However, the facility does not have the capacity to capture the resulting energy yet and as such, the produced biogas is flared into the atmosphere. A biogas capture and upgrading project is underway at this site, and the findings from this study are envisaged to provide baseline empirical evidence and information relating to the environmental implications of such upgrades. All visited facilities preferred anonymity, but their inventory data is provided separately as supplementary material (Table A1-Appendix A). Expert knowledge on biogas production was provided by members of the Canadian Biogas Association (CBA) who validated the final inventory data of the representative reference site. Since at the time of the study there was no AD-RNG facility in Ontario (London Inc. magazine, 2021), the RNG upgrading technology inventory data was modelled based on a recent study undertaken by Zhang et al.(2020), according to a chemical amine scrubbing process, which consists of absorbing CO2 and H2S from biogas into a liquid phase using a mono-ethanolamine solution (biogas 1.48 m³ upgrades to 1 m³ biomethane).

The EcoInvent 3.6 updated database was used for contextualizing the inventory and the commercial LCA software, SimaPro version 9.1.1, was used for LCA analysis. The Tool for the Reduction and Assessment of Chemical and other environmental Impacts (TRACI) developed by the U. S. Environmental Protection Agency has been selected as a midpointoriented life-cycle impact assessment (LCIA) methodology and the following impact categories were selected: Ozone depletion, Global warming, Smog, Acidification, Eutrophication, Carcinogenic, noncarcinogenic, Respiratory effects, Ecotoxicity, and Fossil fuel depletion. The TRACI methodology is consistent with North American locations including Canada, and reflects the state of developments (Bare,



Fig. 1. System boundary of the study (the transportation of the SSO to the AD facility is not included in the analysis).

2011), consistent with EPA regulations and policy as well as best-available practice for LCIA in the United States (Hischier et al., 2010). Inventory data are shown in Table 1, showing inputs and outputs of the averaged representative AD plant, followed by separate biogas co-generation, and upgrading systems (AD-CHP, and AD-RNG) (see Table 2).

3. Results

The following section describes the results of the analysis of the investigated representative AD system which consists of two biogas upgrading pathways: (1) AD-CHP for heat and electricity production or (2) AD-RNG through chemical scrubbing using an amine-based solution. For an improved environmental performance, the environmental burdens and hotspots that characterize each production step are identified. According to the LCA impact assessment procedure, all impacts are classified to the impact categories, then characterized and eventually normalized according to conversion factors available within the TRACI 2.1 method. The resulting normalized and dimensionless indicator values give the ranking of the analyzed system within the defined reference for the respective impact categories. Thus, the aim of normalizing the indicator results is to better understand the magnitude for each indicator result of the product system under study (Brentrup et al., 2004; ISO, 2006). Normalization aims to express the results obtained in the characterization phase through dimensionless numerical factors, to allow summing up and comparison between different impact categories. By observing the normalized data, it is possible to understand which impact categories had the greatest environmental load. The normalized impacts using the TRACI 2.1 midpoint method for the AD facility are shown in Fig. 2.

It can be observed that the carcinogenic impact category is the most affected, followed by ecotoxicity, eutrophication, and non-carcinogenic,

Table 1

Inventory data of inputs and outputs of the AD plant to produce biogas from
source separated organic waste (referred to the selected functional unit).

Data	Units	Amount			
Feed to AD plant					
Source Separated Organic waste	Metric t/day	205			
Electricity, medium voltage {Canada without	kWh/MWh _{el}	72.12			
Quebec}					
AD Facility (Supporting infrastructure and equipm	nent) Input				
Concrete (support infrastructure)	dm ³ /MWh _{el}	3.75			
Reinforced steel (equipment)	kg/MWh _{el}	0.10			
Chromium steel (equipment)	g/MWh _{el}	54.46			
Low-alloyed steel (equipment)	g/MWh _{el}	0.28			
Copper (equipment)	g/MWh _{el}	0.84			
High density polyethylene	g/MWh _{el}	34.17			
Polyvinyl chloride	g/MWh _{el}	2.75			
Synthetic rubber	g/MWh _{el}	0.18			
Asphalt (5500 m^2 paved parking area)	kg/MWh _{el}	3.09			
Other steel (building beams, equipment)	g/MWh _{el}	0.71			
Outputs from AD plant					
Biogas (consisting of 64% CH ₄ plus 36% CO ₂)	m ³ /wet tonne	136.39			
	SSO				
Methane (100% CH ₄)	m ³ /wet tonne	87.43			
	SSO				
Carbon dioxide (100% CO ₂)	m3/wet tonne	48.96			
· _	SSO				
End of life waste management (Discussed qualitatively for policy perspectives)					
Digestate	t/yr	95000.00			
Plastics	t/MWhel	0.56			
Glass, grit, eggshells, oysters, etc.	t/MWh _{el}	0.10			
Biofertiliser	t/day	1.64			
Wastewater effluent	t/day	68.33			

Biogenic emissions from the AD plant were not included due to lack of data and were assumed to be generally negligeable.

Wastewater treatment was not included in the LCA, considering that the liquid digestate is directly applied on nearby agricultural farms outside the system boundary.

Table 2

Inventory data of inputs and outputs of biogas co-generation and upgrading options (AD-CHP) and (AD-CHP) referred to the function unit.

AD-CHP unit inputs (Combined heat and power)	Units	Amount			
Lubricating oil	kg∕ MWh _{el}	0.37			
Concrete	dm ³ / MWh _{el}	5,7E-04			
Reinforced steel	kg∕ MWh _{el}	8,1E-03			
Low-alloyed steel	kg∕ MWh _{el}	0.09			
Chromium steel	g/ MWh _{el}	8.37			
Copper	g/ MWh _{el}	0.78			
Polyethylene	g/ MWh _{el}	1.74			
Polyvinyl chloride	g/ MWh _{el}	1.40			
Synthetic rubber	g/ MWh _{el}	0.05			
other steel (building beams, equipment)	kg∕ MWh _{el}	0.13			
Outputs from CHP					
Electricity output (fed into the electrical grid)	MWh _{el} /per day	61.60			
Heat output (for internal heating needs of digester tanks and buildings)	$\rm MWh_{el}/per~day$	84.37			
0,					
AD-RNG unit inputs (Chemical Amine scrubbing)					
Raw biogas fed into Amine scrubbing plant	m ³	136.39			
Chemical factory, organics	unit	1.00E + 00			
Activated carbon granular	kα	2 71 F-10			

Chemical factory, organics	unit	1.00E + 00
Activated carbon, granular	kg	2.71E-10
Water, deionized	kg	5.00E-04
Monoethanolamine	kg	3.00E-02
Activated silica	kg	3.00E-05
Electricity, medium voltage	kWh	1.28E-02
Outputs from AD-RNG		
Renewable natural gas	m ³ /per wet	92.30
	tonne SSO	
Emissions: kg/136.39 m ³ biogas at 64% methane.		
Methane, non-fossil	kg	5.19E-03
Hydrogen sulfide	kg	6.21E-01
Sulfur dioxide	kg	8.12E-02
Monoethanolamide	kg	1.94E-04
Ammonia	kg	2.36E-06

*205 tonne wet SSO processed per day generates 61.60 MWhel production per day. This translates into 0.3 MWhel per wet tonne SSO.

respectively. Other impact categories such as ozone depletion and global warming should not be disregarded as they are relevant topical issues with impacts amounting to 2.10E-05 and 2.25E-03 respectively (Ncube et al., 2021). For further interpretation of the environmental profile, the contribution of each input to the same impact category is also shown. Figs. 3–5 show characterized impacts of the AD and the two-biogas co-generation and upgrading options, namely AD-CHP, and AD-RNG.

The input with the highest overall contribution (above 60%) to all the impact categories is electricity which is derived from the grid. The environmental load of the electricity is attributed to the production, marketing, and distribution of the medium voltage electricity for Canada, excluding Quebec. Additionally, the infrastructure supporting the construction of the AD facility consists of steel, concrete, and asphalt which are considered in respect of their function, weight, and lifespan. For example, steel production has a significant environmental burden in the carcinogenic impact category by taking up 50% of the total generated impact, due to the formation of carcinogenic substances during its production and distribution. In the same logic, asphalt covering 5500 m^2 of paved driveway area with a 30yr life span, has a higher environmental load in the ozone depletion and fossil resource depletion impact categories. The compartments contributing to ozone depletion such as Ethane-trichloro, HCFC-140 in the background dataset along the supply chain based on the production and marketing of asphalt are responsible for the significant environmental burden. On the other hand, copper wiring and polyplastics are responsible for generating insignificant environmental loads on all the impact categories, contributing less than 4% of the total impact.

When the biogas has been produced through the anaerobic digestion process, it is further transferred to the corresponding cogeneration unit



Fig. 2. Normalized impacts of the anaerobic digestion of SSO (referred to the selected functional unit).

(CHP). The CHP unit consists of an internal combustion biogas-powered engine that drives an electrical generator to produce electricity and heat. The produced electrical power is sold to the local electrical utility company and the heat is used internally as thermal energy for process and space heating purposes. Fig. 4 shows and describes the characterized environmental impacts of co-generating electricity and heat at the AD-CHP unit. As the environmental burden of biogas is carried forward to the AD-CHP unit, 90% of the environmental burden is attributed to the previous anaerobic digestion process. In addition, since the CHP facility has engines, lubricating oil is commonly used for cooling purposes and to reduce friction, causing 18% of the impact on the ozone depletion impact category and 15% on the fossil fuel depletion impact category. The CHP engines require oil changes every 2000 operating hours. For a facility that runs throughout the year, this oil change frequency equates to 4.4 oil changes per year, making lubricating oil a major hotspot input material. Steel as a supporting material input to the structure of the CHP facility, results in a noticeable environmental burden on the carcinogenic impact category due to its production and distribution along the supply chain. Other inputs, such as plastics and concrete, can be considered as insignificant hotspots at the AD-CHP unit.

Alternatively, instead of co-generating heat and electricity, the biogas can potentially get upgraded to renewable natural gas, also called "biomethane". The concurrent generation of heat, electricity, and renewable natural gas can be regarded as more beneficial, but this depends on the financial capacity to do so by any bioenergy project proponent. As with the previous logic in Fig. 4, the AD-RNG extended system using the amine chemical scrubbing biogas upgrading technique, represents an opportunity to upgrade biogas into biomethane. The biogas continues to carry the environmental burdens from the AD process and as expected, the impacts associated with biogas production remain significantly higher compared to the other input materials added at the RNG production unit.

It is worth noting that the biogas upgrading process also releases local biogenic emissions of CO₂, sulphides, and other pollutants affecting the following impact categories: global warming (3.77E-1 kg CO₂ eq); acidification (4.67E-2 kg SO₂ eq) and respiratory effects (3.67E-3 kg PM 2.5 eq). The environmental load of silica and electricity input flows is also evidenced; however, the generated impacts are insignificant with a contribution less than 10% in all the investigated impact categories.

Once the contribution of each considered process to the overall environmental load of the investigated system has been described, a comparison of the co-generation and upgrading options can be undertaken to inform decision-makers (Table 3). Furthermore, the avoided production of fossil-based electricity, natural gas, fossil-based heat, and inorganic fertilizer can be compared against their renewable counterparts (Table 4) (see Table 5).

The characterized values in Table 3 compare the impacts generated from AD-CHP to AD-RNG alternatives. It is evident that the AD-CHP option has better environmental performance compared to the AD-RNG route in all the analyzed environmental impact categories.

To account for the avoided burdens of producing non-renewable energy and chemical fertilizer, Table 4 gives some insights. For example, by deepening the focus on carcinogens (comparative toxicity unit for humans), ecotoxicity (comparative toxicity unit for ecotoxicity), eutrophication impact categories (the most affected impact categories in Fig. 2), global warming, and fossil fuel depletion impact categories (considered due to their topical nature in sustainability studies), the generated renewable co-products have significantly lower environmental impacts compared to their fossil derived counterparts and this confirms the much-touted environmental benefits of shifting towards food and organic waste for producing renewable energy as well as organic fertilizers (Santagata et al., 2020a,b). According to Wang et al. (2021) an integrated AD-CHP scenario can reduce global warming



Fig. 3. Characterized impacts of the anaerobic digestion of SSO (referred to the selected functional unit).

potential (GWP) by 1060-1290 kg CO2-eq per dry tonne waste processed, due to the benefits emanating from the substitution of fossil-derived energy, chemical fertilizers, and direct biogenic emission capturing. This GWP reduction is further confirmed by findings from Whiting and Azapagic (2014), highlighting a 50% lower GWP of using renewable energy from AD of waste than from fossil derived energy sources. Furthermore, Kelly et al. (2014) recommended that installing a CHP plant unit alongside anaerobic digestion can result in significant reductions in GHG emissions, confirming significant energy and carbon savings as has also been highlighted by findings from this present study. The calculated impacts of the fossil counterparts were derived from the EcoInvent 3.6 database showing the production of electricity, medium voltage {CA-ON}, natural gas {CA-QC}, heat, district, or industrial, natural gas {CA-QC} and phosphate fertilizer, as P2O5 {CA-QC}. In Table 4, the corresponding substitution of fossil resource flows with renewable sources (namely renewable electricity, renewable natural gas, renewable heat and biofertiliser) generates negative values. These negative values indicate a reduction of the impacts derived from the substitution of fossil-derived material with renewables, while positive values indicate an increase in the impacts. The minor positive values comparing RNG and fossil-derived natural gas in the ecotoxicity, eutrophication and global warming impact categories, indicate areas of concern as raised by Blum et al. (2020) in connection to increased circularity. Renewable energy production and provision should be approached with caution as the entire supporting infrastructure and background production processes are not entirely composed of renewable materials (Zucaro et al., 2017). For example, electricity and steel have been identified as hotspot input flows in the anaerobic digestion process and lubrication oil in the AD-CHP unit among other non-renewable materials such as concrete, and asphalt that suppor the infrastructure and needed processes. This need for non-renewable inputs

highlights the challenge faced in implementing renewable energy/material production and circular economy, and such developments may not necessarily equate to greater sustainability (Figge et al., 2014; Harris et al., 2021). However, from the overall results in Table 4, the benefits (negative values) of substituting fossil-derived materials with renewables can be appreciated. In particular, fossil depletion and global warming impact categories have highlighted significant gains with much higher negative values compared to other impact categories. Table 4 shows the selected impact categories which were compared to highlight the benefits of avoiding the use of fossil derived energy sources.

3.1. Uncertainty analysis

Due to several factors including quality of data, assumptions made, system boundaries and methods used for impact assessment which affects the overall results of the LCA study, uncertainties still exist (Cellura et al., 2011). Furthermore, uncertainties are related to factors that affect data quality, such as variations in the composition of SSO feedstock material that can influence the yield of biogas and the performance of the AD facility in the attributional LCA (Bartocci et al., 2020). Against this background, a Monte Carlo uncertainty assessment at a 95% confidence interval is performed using the SimaPro software to test the reliability and robustness of the results. Table 5 shows the results of the selected midpoint impact indicator categories related to upgrading biogas via AD-RNG (A) and AD-CHP (B), indicating mean, standard deviation (SD), coefficient of variation (Cv, defined as the ratio between the SD and the mean), standard error of the mean (SEM, defined as the standard deviation of the sampling distribution of the mean). Cv ranges of 5-51% are regarded as lower variations and this included the following impact categories: Acidification, Fossil fuel scarcity, Global



Fig. 4. Characterized impacts of the cogeneration of heat and electricity (CHP) (referred to the selected functional unit).

warming, Ozone depletion, Respiratory effects and Smog, thus confirming that the results in these categories have good reliability (Beccali et al., 2010). Ecotoxicity and Eutrophication are slightly above 51%, whereas Carcinogenic impact categories show relatively higher uncertainty ranges above 100%, thus calling for further checks and studies on more localised, specific data over longer periods in order to reduce uncertainty linked to this impact category (Florio et al., 2019).

4. Discussion

The SSO waste has been processed and treated to produce biogas where electricity and steel have been identified as critical hotspots in the anaerobic digestion process. It is in the best interest of the AD operator to use the renewable electricity that is already produced on site instead of relying on the grid. Due to economical and contractual obligations signed between the producers and the regulatory authority through the Ontario Feed-In Tariff (FIT) program, this is, however, impossible despite the highlighted environmental benefits.

Due to similarities in functional unit and some assumptions made, it is possible to compare the results of this study (AD-CHP) with those achieved by Wang et al. (2021) on a study carried out in British Columbia Edwards et al. (2017). The overall global warming potential relative to the processing of 1 wet metric tonne of SSO in the studied AD-CHP and AD-RNG system fragments are 11.8 kg CO₂ eq and 29.6 kg CO₂ eq, respectively. About 205 tonnes of wet SSO is processed per day, resulting in 2.42 t CO₂ per day from the AD-CHP unit and 6.06 t CO₂ per day from the AD-RNG upgrade system. Wang et al. (2021) estimated 115 t/month of CO₂ eq from the processing of 350 dry mass tonnes per day of food waste, and our study approximates 72.57 t/month of CO₂ eq at the AD-CHP unit and 182.04 t/month CO₂ eq at the AD-RNG upgrade system. The study by Wang et al. (2021) used a case study in British Columbia, Canada, making it more reliable and comparable with our study. Assuming the British Columbian site processes the same amount of SSO per day (205 t/day as in the study inventory), the total carbon emissions will be equivalent to 67.36 t/month CO2 eq, making it closer to our results of 72.57 t/month of CO2 eq (AD-CHP). During a study in Australia, Edwards et al. (2017) recorded 6360 t CO2 eq per 91,311 metric tonnes of treated organic waste per year. It is, however, generally difficult to compare the results of the performance of AD-CHP and AD-RNG units with other studies due to dissimilar functional units and the type of feedstock biomass materials (Hijazi et al., 2016; Florio et al., 2019). Some researchers have already investigated different biogas upgrading options, such as membrane separation, cryogenic separation, pressure swing adsorption, chemical scrubbing, and high pressure water scrubbing technologies against the CHP option and have concluded that CHP performs better from an environmental point of view (Florio et al., 2019), thus confirming our findings from an environmental point of view (Wang et al., 2021).

The AD-CHP system performed better from an environmental point of view with 11.8 kg CO_2 eq when compared to the AD-RNG with 29.6 kg CO₂ eq biogas upgrade. The difference is more than two times, warranting some negative environmental feedbacks which may dampen the adoption of AD-RNG upgrading technologies. However, in view of the increasing attention towards RNG in Canada, both upgrading systems (AD-CHP and AD-RNG) can be integrated in such a way that they can operate simultaneously to increase economic benefits. At the time of the study, there were no major commercial RNG upgrading projects in Ontario, Canada. Only recently (less than 6 months) did the StormFisher RNG upgrade come into full operation, becoming one of the few RNGproducing facilities in North America (London Inc. magazine, 2021). Against this background, further studies on cost benefit analysis are recommended, to provide deepened insights on economic performance



Fig. 5. Characterized impacts of the chemical scrubber biogas upgrading system (referred to the selected functional unit).

Table 3

Comparing the relative contribution of the characterized impacts of biogas upgrading technologies ((AD-CHP and AD-RNG) based on processing 1 wet metric ton of SSO).

Impact category	Unit	CHP	RNG	
Ozone depletion	kg CFC-11 eq	1.10E-06	1.73E-06	
Global warming	kg CO ₂ eq	1.18 E+01	2.98 E+01	
Smog	kg O ₃ eq	6.76E-01	1.24 E+00	
Acidification	kg SO ₂ eq	6.45E-02	1.71E-01	
Eutrophication	kg N eq	1.51E-01	2.62E-01	
Carcinogenics	CTUh	3.01E-06	4.78E-06	
Non carcinogenics	CTUh	6.86E-06	1.20E-05	
Respiratory effects	kg PM2.5 eq	1.13E-02	2.24E-02	
Ecotoxicity	CTUe	2.50 E+02	4.17 E+02	
Fossil fuel depletion	MJ surplus	1.52 E + 01	2.54 E+01	

which was beyond the scope of this study. According to a recent report published by the Canadian Biogas Association (2021), the clustering of RNG projects in collaboration with multiple AD agricultural farms is theoretically capable of generating and injecting approximately 6 Petajoules (PJ) of RNG into the natural gas grid. This clustering of RNG projects is a means of managing Ontario's food and organic waste problem.

According to Leonzio (2016), despite the highest upgrading efficiency using the mono-ethanolamine solution during chemical scrubbing, some environmental and technical improvements are still needed. In addition, pre-existing conditions at specific sites, economic performance and market strategies should be taken into consideration when selecting biogas upgrading technologies (Ardolino et al., 2020, Eggemann et al., 2020). For instance, in a geographic setting like British Columbia, Canada, the capital cost of an AD system ranges from \$690– \$–1060 (Canadian dollar) per tonne dry matter (tDM) processed per year (Wang et al., 2021). Additionally, biogas valorisation through combustion to heat and electricity (AD-CHP unit) and connecting to the grid incurs an additional 15–19% of total capital costs. Likewise, purification of biogas towards biomethane will incur 9–16%, respectively. Furthermore, other operating costs such as labour, feedstock cost, pipeline costs, depreciation, and loan interests are expected to increase production costs. Coupled with lower natural gas prices (RNG prices in Canada of up to \$30/GJ are currently being offered by gas utilities) and the absence of financial support from government, AD operators often rely on tipping fees and the sale of biofertiliser as extra revenue streams in order to balance the costs of production and capital costs. These additional revenue streams highlight the importance of pursuing a circular economy to obtain and recover value-added by-products, even from plastic residuals and wastewater.

4.1. Circular and policy outlooks on plastics and wastewater from AD facilities

Stepping away from LCA and into perspectives for further discourse relating to increasing recycling and circularity within AD facilities, the handling and treatment of wastewater and plastics, within the studied system is discussed for increased environmental awareness and informing policy. For example, growing concern over the environmental damage associated with conventional product packaging has led to keen interest in sustainable packaging (Herbes et al., 2018). However, there is a general lack of a coordinated approach to addressing the current plastics challenge in Canada, including at biogas plants. For example, about 14%–17% of the raw feedstock arriving at the AD facility is composed of plastics, which are subsequently landfilled at the studied sites. There is a significant increase in the transition of commercial food providers from plastic-based food packaging, containers, and ancillary items to those purporting to be "compostable" or "biodegradable",

Table 4

Comparison of selected characterized impacts of the production of renewables and their fossil derived counterparts.

Impact category	Carcinogenics	Ecotoxicity	Eutrophication	Global warming	Fossil fuel depletion
Unit	CTUh ^a	CTUe ^b	kg N eq	kg CO2 eq	MJ surplus
Electricity from AD-CHP	2.19E-06	1.81 E+02	1.09E-01	1.18 E+01	1.10 E+01
Electricity from Grid {CA-ON}	5.02E-07	6.46 E+01	2.18E-02	1.97 E+01	3.87 E+01
(+/-)	0.77	0.64	0.80	-0.67	-2.51
Renewable Natural Gas (AD-RNG)	4.75E-06	4.15 E+02	2.61E-01	2.98 E+01	2.53 E+01
Natural gas-High pressure {CA-AB}	1.98E-07	1.89 E+01	2.44E-02	3.09 E+00	3.38 E+02
(+/-)	-0.04	0.95	0.91	0.90	-12.38
Heat from AD-CHP	3.00E-06	2.49 E+02	1.50E-01	1.62 E+01	1.51 E+01
Heat-Industrial {CA-QC}	1.40E-07	1.52 E+01	1.57E-02	9.57 E+01	2.30 E+02
(+/-)	-0.05	0.94	0.90	-4.90	-14.22
Biofertiliser from AD plant	4.68E-06	3.93 E+02	2.55E-01	2.70 E+01	2.22 E+01
Phosphate fertiliser- {CA-QC}	4.94E-05	1.59 E+04	1.83 E+00	3.15 E+02	3.71 E+02
(+/-)	-10.56	-39.29	-6.18	-10.69	-15.73

^a Comparative Toxicity Unit for humans.

^b Comparative Toxicity Units for ecotoxicity.

Table 5

Results displaying Monte Carlo uncertainty analysis of AD-RNG (A) vs AD-CHP (B) in a table at 95% confidence interval.

Impact category	$\begin{array}{c} A \geq \\ B \end{array}$	Mean	Median	SD	CV %	SEM
Acidification	100	1.06E-	1.06E-	3.12E-	2.95	9.88E-
		01	01	03		05
Carcinogenics	100	1.84E-	1.17E-	6.65E-	360.89	2.10E-
		06	06	06		07
Ecotoxicity	100	1.64	1.45	9.37	57.25	2.96
		E+02	E+02	E+01		E+00
Eutrophication	100	1.07E-	8.95E-	6.50E-	60.57	2.06E-
		01	02	02		03
Fossil fuel	100	1.02	1.01	8.42E-	8.29	2.66E-
depletion		E+01	E+01	01		02
Global warming	100	1.34	1.34	8.98E-	6.70	2.84E-
		E+01	E+01	01		02
Non	64.1	5.21E-	4.56E-	1.46E-	280.27	4.61E-
carcinogenics		06	06	05		07
Ozone depletion	100	6.37E-	5.69E-	2.64E-	41.39	8.33E-
		07	07	07		09
Respiratory	100	1.11E-	1.10E-	4.30E-	3.88	1.36E-
effects		02	02	04		05
Smog	100	5.63E-	5.62E-	3.12E-	5.53	9.86E-
		01	01	02		04

which governments in Canada have continued to vaguely endorse without adopting any standards as to their material requirements or performance. AD facilities are designed to remove plastics through physical and mechanical means, not distinguishing between compostable and conventional plastics, all of which are then directed to landfill.

These plastic materials threaten the sustainability of biogas plant operations due to the environmental footprint associated with producing the bioplastic material which is subsequently landfilled. As such, there is an existing technical and methodological divide between the end-of-life design for a significant portion of the food waste stream and its actual recovery which calls for action towards sustainable food packaging using biodegradable materials compatible with the intended destination, be that aerobic composting or anaerobic digestion. According to Shrestha et al. (2020), bioplastics are commonly favoured as an alternative to single use fossil-based plastics. In addition, due to their biodegradable properties, these bioplastics are expected to yield additional benefit of biogas after their use. Shrestha et al. (2020), however, caution on the biodegradability time of these bioplastics which far exceeds the processing times in anaerobic digestion facilities. Canada does not currently have any national bioplastics content standards and therefore, more research is viewed, as necessary. Further, based upon international disagreement on this issue, it is not clear what standard would be introduced in Canada, if any, to denote when a plastic is

"biodegradable" or "compostable" material and the regulatory trends suggest that local processing infrastructure capacities and not international product content standards will determine which, if any, bioplastics are ultimately permitted in Ontario or Canada (Government of Canada, 2021; Ministry of Agriculture, 2021).

To further realize the full benefits within a circular perspective in organic waste processing and treatment, the provincial and federal governments need to set a clear path forward and a strategy with longterm goals on the wastewater end usage. This strategy is necessary to facilitate the management and treatment of wastewater. This discussion will provide the framework for potential wastewater treatment pathways. While efforts to-date are noteworthy and important, such as farm spreading, they have largely been uncoordinated and have not created the policy drivers necessary for greater progress. For the nominal site examined in this study the disposition of the digested organic slurry, called digestate, is an important aspect. Because digestate is a watery slurry, containing more than 90% water, there are three general approaches to managing this residual material: the digestate slurry can be i) stored and seasonally land-applied as a liquid fertilizer, ii) filtered or otherwise separated into a water-based and a solid fertilizer, and iii) filtered or otherwise separated to produce a solid fertilizer and then process the watery residual in a wastewater treatment process before discharging the cleaned water to the natural environment. All three approaches are currently being used in Ontario. In the model facility used in this study we have taken the first approach: the digestate slurry is stored and seasonally applied directly to agricultural land without further processing. This approach minimizes the equipment and energy inputs to the system, based on the assumption that the digestate does not need to be transported a significant distance to reach the agricultural lands. This assumption is not always correct, as in some instances, AD facilities are located in urban areas and may need to transport the digestate a significant distance. This direct land application approach also retains the beneficial nitrogen and phosphorus nutrients in the digestate and returns them to the land following the circular economy approach.

The second approach, separating the liquid and solid digestate and managing these two residual streams separately, has benefit where the recovered solids have value. For example, on-farm digesters can recover undigested fibre from the digestate for use in animal bedding. The final approach involves separating the solid and liquid portion of the digestate and managing each stream separately. In Ontario, the facilities that do this typically send the wet digested solids to be further processed by aerobic composting, while the watery portion is processed in wastewater treatment facilities before being discharged into the natural environment. Managing the two solid and liquid streams in this way affects the circular nature of the process. The ammonia nitrogen is predominantly present in the watery portion; a major objective of the wastewater treatment process is to convert this ammonia nitrogen to i) nitrogen gas, which is discharged into the air, or ii) nitrate which is discharged into the lakes or river with the treated water. In neither case is the nutrient value of the ammonia nitrogen in the digestate captured for beneficial use.

5. Conclusions

The proper processing of organic waste offers environmental and economic opportunities in a future with restrictions upon fossil-based fuels and materials. The environmental burden of an AD facility was evaluated considering all the different stages in the processing and treatment of organic waste at an averaged representative AD facility in Ontario, Canada.

Utilizing the standardized LCA method for environmental assessments, the options of (i) recovering biogas in a co-generation unit (AD-CHP) to produce heat and electricity and (ii) upgrading it to biomethane (AD-RNG) were explored. As a result, electricity and steel were identified as critical hotspot inputs carrying a significant environmental burden in the anaerobic digestion process. Moreover, lubrication oil used in the CHP unit was identified as a dominant hotspot input flow for the AD-CHP system, while activated silica and electricity were highlighted as the major hotspot input flows in the chemical amine scrubbing for AD-RNG system.

A comparison of the two biogas co-generation and upgrading alternatives concluded that, the AD-CHP plant generates a much lower environmental load when compared to the AD-RNG unit. The avoided burden of producing fossil-based electricity, natural gas, and chemical fertilizer was also included. Biobased heat, electricity, and renewable natural gas, plus biofertiliser, indicated better environmental performance with significant reductions in CO₂ equivalent emissions and in the depletion of fossil resources. In conclusion, based on the treatment and processing of source separated organic waste at an AD facility, the effort to divert organic waste from landfills brought reductions in the generated environmental impacts. The life cycle-based results have provided insights to inform policy directions towards the much-needed cooperation among researchers, regulators, and other involved stakeholders in developing coherent approaches in pursuing organic waste management and biogas upgrading alternatives. In addition, the study recommends some further research on cost and benefit analysis for each biogas upgrading technology to achieve more insights from an economical point of view.

Credit author statement

Amos Ncube: Conceptualization, Investigation, Formal analysis, Validation, Original draft preparation, Writing - Reviewing & Editing; Jonathan Cocker: Supervision, Validation, Writing - Reviewing & Editing; David Ellis: Data curation, Formal analysis, Investigation, Validation, Writing - Reviewing & Editing; Gabriella Fiorentino: Supervision, Project administration, Validation, Writing - Reviewing & Editing.

Declaration of competing interest

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

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Appendix A. Supplementary data

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