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Decarbonizing anaerobic digestion in Canada's wastewater resource recovery facilities: an opportunity to attain carbon-neutral biogas production and its potential to offset waste management sector carbon emissions

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Anaerobic digestion (AD) is a process for valorizing sewage sludge through biogas production from sludge that is processed in water resource recovery facilities (WRRFs). Biogas can be combusted to generate thermal and electrical energy, converting the methane component of biogas into near-neutral carbon dioxide emissions of biogenic origin. Sewage-derived biogas is comprised of ≈98% biogenic carbon as methane and carbon dioxide. The capture of carbon dioxide of biogenic origin offers an opportunity to generate negative carbon emissions to offset the greenhouse gas (GHG) emissions from biogas production and utilization processes. In this work, an inventory of estimated GHG releases from AD system emissions and leaks in WRRFs across Canada was estimated with open-source data. The potentials to deploy different carbon capture processes to achieve carbon-neutral biogas systems were then estimated. The results show that implementing carbon capture systems downstream from biogas utilization or in biogas upgrading units could offset the carbon footprint of biogas generation systems. Implementing in-digester carbon capture would result in partial net GHG emission reduction. Moreover, some carbon capture processes would result in a surplus of negative GHG emissions with potential to offset the GHG emissions from other waste management activities, such as incineration.

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Sustainability spotlight

Biogas produced in wastewater resource recovery facilities (WRRFs) is comprised of 98% biogenic carbon. The capture of biogenic-origin carbon dioxide downstream from biogas utilization or in biogas upgrading units generates negative carbon emissions that could offset the carbon footprint of methane leaks and achieve effective carbon-neutral biogas production and utilization systems. Furthermore, when a surplus of negative carbon emissions is generated, this can be utilized to mitigate greenhouse gas emissions from other anthropogenic activities where the implementation of carbon capture processes faces major challenges or such processes are not yet developed. This work supports several UN Sustainable Development Goals (SDGs), particularly SDG 7 (Affordable and Clean Energy) and SDG 13 (Climate Action).

1 Introduction

Municipal wastewater treatment is evolving towards a more sustainable paradigm, transitioning from the concept of pollution-mitigating wastewater treatment plants (WWTPs) to water resource recovery facilities (WRRFs) by considering the wastewater treatment process as a resource for energy, nutrients, and sanitized water.[1] Here, WRRFs refer to WWTPs that implement anaerobic digestion (AD) units for sewage sludge treatment, producing biogas and digestate. Biogas can be used as a source of electricity and/or thermal energy, while the digesate obtained can be used in croplands as a soil amendment and a source of nutrients.[2] The major components of biogas generated by anaerobic digestion of sewage sludge are 50–69 vol% methane and 24–45 vol% carbon dioxide, while minor components include nitrogen (N_2, ≤9 vol%), hydrogen sulfide (H_2S, ≤0.09 vol%), and siloxanes (≤0.004 vol%).[3] Carbon dioxide and methane gases generated by WWTPs and WRRFs are predominantly attributed to modern biogenic sources,[4] *i.e.* the oxidation of organic matter flowing within the natural carbon cycle, instead of fossil carbon which is geologically stored and requires human action to be released to the atmosphere.[5] Therefore, biogenic carbon dioxide emissions will not

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result in a net increase of anthropogenic greenhouse gas (GHGs) emissions to the atmosphere.[6,7] An estimate of carbon dioxide releases from biogas produced from sewage sludge was reported to be 98.4 vol% biogenic and 1.6 vol% fossil carbon.[8] Consequently, marginal fossil carbon emissions can be associated with power and heat production from sewage sludge-based biogas.[9]

However, methane emissions result in net GHG emissions, even if they derive from biogenic sources, since the global warming potential (GWP) over 100 years for methane is 28 times higher than that of carbon dioxide.[10] Although the methane component of biogas is commonly combusted to convert the methane emissions to biogenic carbon dioxide emissions, methane leaks simultaneously occur across the biogas production and utilization processes.[11] These methane leaks contribute to the net emissions of GHG at WRRFs, jeopardizing the sustainability of biogas production as process for the treatment and valorization of sewage sludge.

Biogas production and utilization processes are point sources of carbon dioxide emissions that offer the opportunity to implement carbon capture and storage (CCS) systems to produce negative carbon emissions by capturing biogenic-based carbon dioxide.[5] The negative emissions generated can be used to mitigate the warming effect of methane leaks, and if the negative emissions can offset the methane emissions, operate as a carbon neutral process.[12] Moreover, if the negative emissions created by capturing this carbon dioxide exceed biogas methane emissions from process leaks, the negative emissions surplus can offset the GHG emissions from other anthropogenic activities. This is a promising strategy for sectors facing challenges in implementing zero emissions processes, such as the waste management sector, which in Canada has not achieved a significant reduction of GHG emissions in the last two decades in spite of the continuous implementation of mitigation actions.[13]

Additionally, the reduction of the biogas carbon dioxide concentration can increase biogas energy density, increasing its value as energy carrier.[14] However, carbon capture systems are not commonly implemented in WRRFs, either for capturing the carbon dioxide from biogas combustion off-gases after heat and electricity recovery, nor carbon dioxide separation from methane to upgrade biogas to renewable natural gas (RNG) quality. As a result, the carbon dioxide produced by WRRFs during biogas generation and utilization is not captured and stored, but released into the atmosphere.[15]

We identify two main knowledge gaps to determine whether the production of biogas in WRRFs could result in a carbon-neutral source of energy, *i.e.*, the extent to which CCS systems could contribute to offsetting methane emissions from biogas production leaks remains unquantified.

This work aims to perform an estimation of the carbon capture potential of different CCS systems from the biogas production systems installed in WRRFs in Canada. To achieve this objective, we first present the development of a carbon dioxide emissions inventory from biogas generated by municipal WRRFs in Canada, as there are no public databases collecting this information. Secondly, the carbon dioxide emission estimates informed the CCS potential estimates to abate WRRF GHG emission, and their potential to offset the methane emissions from biogas production and utilization processes. Finally, the contribution of the surplus of negative emissions to offset GHG emissions from other activities within the waste management sector is also assessed to evaluate the contribution of WRRF CCS to the Canadian objective to reach net zero GHG emissions by 2050.[16]

2 Methodology

2.1 Carbon emissions inventory from biogas production and use at Canada's municipal WRRFs

The inventory estimate required identification and collection of public WRRF data, estimation of biogas generation rates for each WRRF, and estimation of the carbon dioxide and methane generation, leaks, and on-site combustion or on-site RNG upgrading. Fig. 1 is a schematic that identifies the mass flow estimates for different biogas use scenarios. With these data, a comparison of the WRRF emissions and carbon capture potential can be made, and binned by Canadian geographical areas.

2.1.1 Identification and collection of open-source data. Relevant data that describe the municipal WRRFs equipped with AD units was obtained from open source data provided by local and provincial government institutions, and verified with a list of facilities provided by Environment and Climate Change Canada.[17] Retrieved data include the locations and magnitudes of the population served, the annual treated wastewater volume, the sludge treatment process types, and the intended biogas use at each WRRF. AD units were reported in all of the Canadian provinces except New Brunswick. The inventory of Canadian WRRFs equipped with AD units can be found in the Zenodo repository reported in the Data availability section.

The Wastewater Systems Effluent Regulations Reported Data is used as inventory of all municipal WRRFs in Canada, and to retrieve the data relative to the water discharged by each facility.[18]

2.1.2 Biogas, methane, and carbon dioxide production rates. The annual biogas production rates are not reported for all of the Canadian WRRFs. Therefore, the annual production rates of biogas, methane, and carbon dioxide were estimated from the wastewater treatment rates and each facility type, as described in eqn (1)–(5). The municipal wastewater treatment capacity of each WRRF is based on the population served and not on the reported treatment design capacity, as this capacity is often designed to handle flow peaks of wastewater that may occur during the year, and thus it does not represent the average annual flowrate.

The annual biosolids mass flow, $\dot{m}_{\text{biosolids}}$, was estimated according to the biosolids generation rate for each type of sludge treatment process, $\text{biosolids}_{\text{generation rate}}$, as shown in eqn (1). The assumed, normalized biosolids generation rates for different sludge treatment processes are listed in Table 1. To account for the uncertainty in the biosolids generation rate, a variation of −22% and +17% was assumed, using an asymmetric triangular distribution.[19]

The biogas mass flow, $\dot{m}_{\text{biogas produced BS}}$, was estimated by eqn (2); where the subscript BS denotes "Baseline Scenario". It

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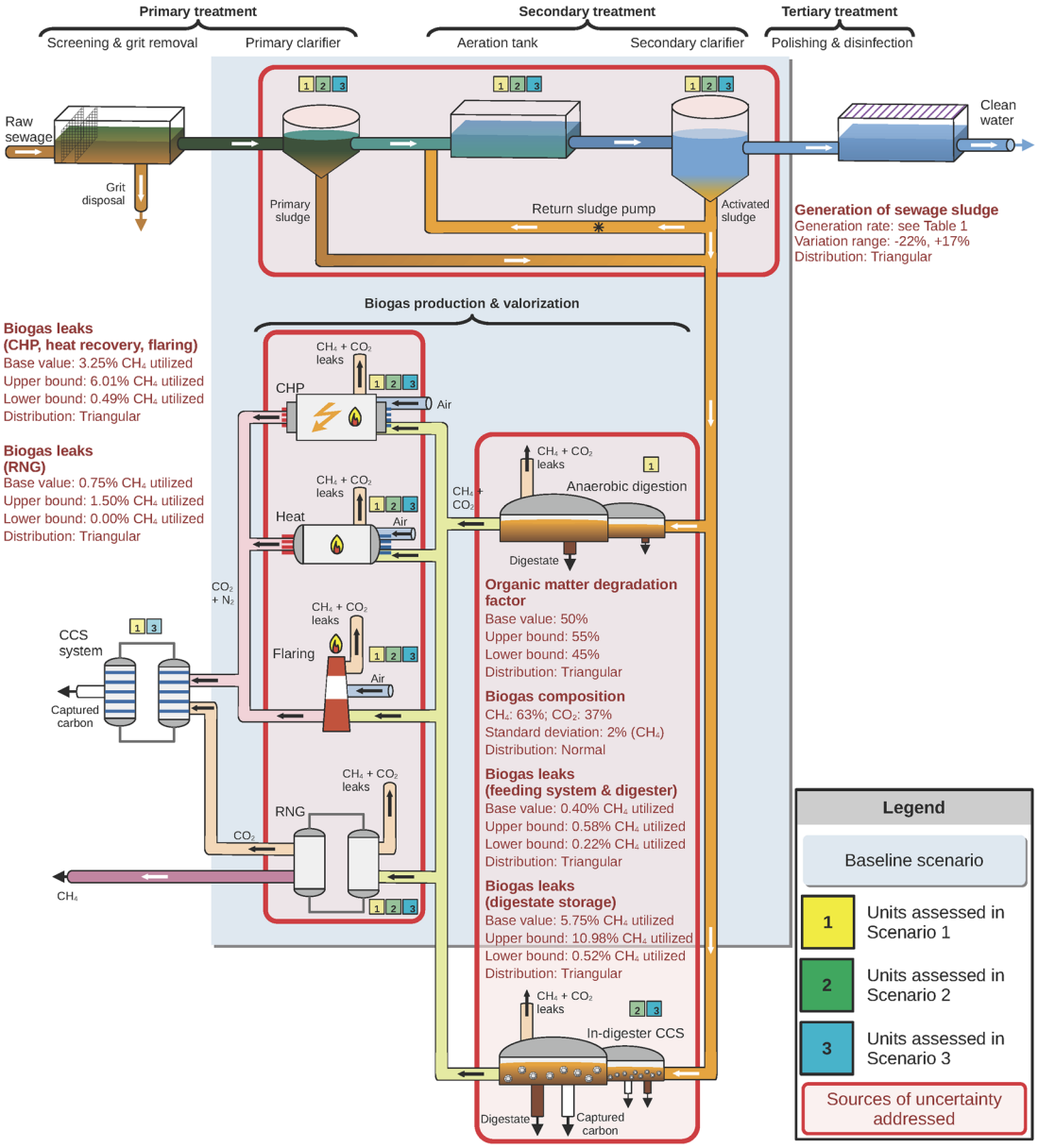

Fig. 1 Carbon capture processes considered and process boundaries for estimating carbon emissions and carbon capture potential.

was assumed that the volatile solids account for 75% of the total solids, that the volatile solids degradation ratio is 55%,[20] and that all biomass degraded is transformed into biogas, so that each kilogram of biomass degraded transformed into one kilogram of biogas.[21] To account for the uncertainty of the biosolids degradation rate, a triangular distribution with a variation of ±10% was assumed.[22]

The biogas composition was assumed to be 63 ± 2 mol% methane and 37 ± 2 mol% carbon dioxide, considering normal distribution.[3] The mass balances of the minor components of biogas were not addressed due to their low mass flows relative to methane and carbon dioxide.

Combining the biogas production and composition, the methane mass flowrate ($\dot{m}_{CH_4\ \text{produced BS}}$) and carbon dioxide mass flowrate ($\dot{m}_{CO_2\ \text{produced BS}}$) were estimated with eqn (3)–(5). MW_i denotes the molecular weight, and x_i denotes the molar fraction of component i.

$$\dot{m}_{\text{biosolids}}\ (\text{kg}_{\text{dry matter}}\ \text{a}^{-1}) = \text{treatment capacity}_{\text{WRRF}}\ (m_{\text{wastewater}}{}^3\ \text{day}^{-1}) \times 365(\text{days a}^{-1}) \times \text{Biosolids}_{\text{generation rate}}\ (\text{kg}_{\text{dry matter}}\ \text{m}^{-3}) \quad (1)$$

$$\dot{m}_{\text{biogas produced BS}}\ (\text{kg a}^{-1}) = \dot{m}_{\text{biosolids}}\ (\text{kg}_{\text{dry matter}}\ \text{a}^{-1}) \times 0.75\ (\text{kg}_{\text{volatile solids}}\ \text{kg}_{\text{dry matter}}{}^{-1}) \times 0.55\ (\text{kg}_{\text{volatile solids degraded}}\ \text{kg}_{\text{volatile solids}}{}^{-1}) \times 1\ (\text{kg}_{\text{biogas}}\ \text{kg}_{\text{volatile solids degraded}}{}^{-1}) \quad (2)$$

$$\text{MW}_{\text{biogas}}\ (\text{kg kmol}^{-1}) = x_{CH_4} \times \text{MW}_{CH_4}\ (\text{kg kmol}^{-1}) + x_{CO_2} \times \text{MW}_{CO_2}\ (\text{kg kmol}^{-1}) \quad (3)$$

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Table 1 Biosolids generation rates for different biosolids treatment processes[19]

Biosolids treatment process	Biosolids generation rate (kg of dry solids per m^3 of wastewater)
Primary sedimentation and conventional activated sludge; Biological nutrient removal or no chemical phosphorus removal	0.180
Primary sedimentation and conventional activated sludge; Chemical phosphorus removal	0.220
Primary sedimentation and conventional activated sludge with anaerobic digestion; Biological nutrient removal or no chemical phosphorus removal	0.115
Primary sedimentation and conventional activated sludge with anaerobic digestion; Chemical phosphorus removal	0.150
Extended aeration; Biological nutrient removal or no chemical phosphorus removal	0.090
Extended aeration; Chemical phosphorus removal	0.120
Extended aeration with aerated sludge holding tank; Biological nutrient removal or no chemical phosphorus removal	0.080
Extended aeration with aerated sludge holding tank; Chemical phosphorus removal	0.110

$$\dot{m}_{CH_4\ \text{produced BS}}\left(\text{kg a}^{-1}\right) = \frac{\dot{m}_{\text{biogas produced BS}}\left(\text{kg a}^{-1}\right)}{\text{MW}_{\text{biogas}}\left(\text{kg kmol}^{-1}\right)} \times x_{CH_4} \times \text{MW}_{CH_4}\left(\text{kg kmol}^{-1}\right) \quad (4)$$

$$\dot{m}_{CO_2\ \text{produced BS}}\left(\text{kg a}^{-1}\right) = \frac{\dot{m}_{\text{biogas produced BS}}\left(\text{kg a}^{-1}\right)}{\text{MW}_{\text{biogas}}\left(\text{kg kmol}^{-1}\right)} \times x_{CO_2} \times \text{MW}_{CO_2}\left(\text{kg kmol}^{-1}\right) \quad (5)$$

2.1.3 Biogas leak rates. Biogas leaks during its production and utilization are especially relevant because these direct methane emissions absorb 28 times more infrared thermal radiation than carbon dioxide over a time span of 100 years.[10] Since the fraction of methane leaks at each process step relative to the flow of methane valorized is reported in the literature, the estimated biogas leaks at each process step are based on estimates of the methane leaks. First, the fraction of methane leaked at each process step, κ_{CH_4}, were combined with the estimate of methane produced to calculate the mass flow of methane used in the valorization process of each WRRF, $\dot{m}_{CH_4\text{used BS}}$, as shown in eqn (6). Secondly, valorized biogas flow, $\dot{m}_{\text{biogas used BS}}$, was estimated in eqn (7) by combining the methane used in such step and the methane content in biogas. Third, the mass flow of methane leaked at each process step, $\dot{m}_{CH_4\ \text{leaked BS}_{\text{unit}}}$, was estimated as the product of the methane used in the valorization stage, and the fraction of methane leaked in this step, as shown in eqn (8). Finally, the mass flows of carbon dioxide and biogas leaked at each process step, $\dot{m}_{CO_2\ \text{leaked BS}_{\text{unit}}}$ and $\dot{m}_{\text{biogas leaked BS}_{\text{unit}}}$ respectively, were estimated in eqn (9) and (10) combining the flow of methane leaked at such step and the composition of biogas. The fraction of methane leaks at each process step and their variability are collected in Table 2. A triangular distribution was assumed for these parameters.

$$\dot{m}_{CH_4\ \text{used BS}}\left(\text{kg a}^{-1}\right) = \frac{\dot{m}_{CH_4\ \text{produced BS}}\left(\text{kg a}^{-1}\right)}{1 + \sum_{\text{unit}} \frac{\kappa_{CH_4\ \text{unit}}(\%)}{100}}; \quad (6)$$

unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

$$\dot{m}_{\text{biogas used BS}}\left(\text{kg a}^{-1}\right) = \frac{\dot{m}_{CH_4\ \text{used BS}}\left(\text{kg a}^{-1}\right)}{\text{MW}_{CH_4}\left(\text{kg kmol}^{-1}\right) \times x_{CH_4}} \times \text{MW}_{\text{biogas}}\left(\text{kg kmol}^{-1}\right) \quad (7)$$

$$\dot{m}_{CH_4\ \text{leaked BS}_{\text{unit}}}\left(\text{kg a}^{-1}\right) = \dot{m}_{CH_4\ \text{used BS}}\left(\text{kg a}^{-1}\right) \times \frac{\kappa_{CH_4\ \text{unit}}(\%)}{100}; \quad (8)$$

∀ unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

$$\dot{m}_{CO_2\ \text{leaked BS}_{\text{unit}}}\left(\text{kg a}^{-1}\right) = \frac{\dot{m}_{CH_4\ \text{leaked BS}_{\text{unit}}}\left(\text{kg a}^{-1}\right)}{\text{MW}_{CH_4}\left(\text{kg kmol}^{-1}\right)} \times \frac{x_{CO_2}}{x_{CH_4}} \times \text{MW}_{CO_2}\left(\text{kg kmol}^{-1}\right); \quad (9)$$

∀ unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

$$\dot{m}_{\text{biogas leaked BS}_{\text{unit}}}\left(\text{kg a}^{-1}\right) = \frac{\dot{m}_{CH_4\ \text{leaked BS}_{\text{unit}}}\left(\text{kg a}^{-1}\right)}{\text{MW}_{CH_4}\left(\text{kg kmol}^{-1}\right) \times x_{CH_4}} \times \text{MW}_{\text{biogas}}\left(\text{kg kmol}^{-1}\right); \quad (10)$$

∀ unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

2.1.4 Onsite biogas valorization emission rates. Total methane combustion was assumed for those biogas applications that have on-site combustion, *i.e.* heat and electric power cogeneration, heat recovery, or biogas flaring. Thus, the carbon dioxide emissions of these processes, $\dot{m}_{CO_2\ \text{released biogas combustion or upgrading BS}}$, were assumed to be the sum of the biogas carbon dioxide component and the carbon dioxide generated by biogas methane combustion, with an assumed 1 : 1 molar ratio of carbon dioxide generation after methane combustion, as shown in eqn (11). Separation of biogas into carbon dioxide and methane streams, known as biogas upgrading, produces renewable natural gas (RNG). It is assumed that the RNG was transported and combusted outside of the WRRF battery limits, which lie outside of the study boundaries, as it is shown in Fig. 1. Although the carbon dioxide separated from biogas can be captured, it is commonly emitted to the atmosphere. As

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Table 2 Fraction of methane leaks at each process step relative to the flow of methane valorized.[11] The base and uncertainty values are reported in the same basis

Unit	Fraction of methane leaked relative to the methane used in the valorization stage ($\kappa_{CH_4\ unit}$)
Digester and feeding system	0.4% ± 0.18%
Digestate storage	5.75% ± 5.23%
Combustion-based valorization processes (CHP, heat recovery, and flaring)	3.25% ± 2.76%
RNG production	0.75% ± 0.75%

such, for this study the separated carbon dioxide was assumed to be emitted.[23]

2.1.5 Total GHG emission rate. The total estimated GHG emissions from biogas production and utilization without carbon capture systems, $\dot{m}_{CO_2\ \text{released biogas combustion or upgrading BS}}$, were estimated with eqn (12), where GWP_{CH_4} denotes the global warming potential over 100 years of methane, valued at 28 CO_{2eq},[10] and CO_{2eq} is defined as the equivalent amount of carbon dioxide that would result in the same temperature change over the given time horizon.[24]

$$\dot{m}_{CO_2\ \text{released biogas combustion or upgrading BS}}\left(\text{kg a}^{-1}\right) = \dot{m}_{CO_2\ \text{produced BS}}\left(\text{kg a}^{-1}\right) - \sum_{\text{unit}} \dot{m}_{CO_2\ \text{leaked BS}_{\text{unit}}}\left(\text{kg a}^{-1}\right) + \left[\dot{m}_{CH_4\ \text{used BS}}\left(\text{kg a}^{-1}\right) \times \frac{MW_{CO_2}\left(\text{kg kmol}^{-1}\right)}{MW_{CH_4}\left(\text{kg kmol}^{-1}\right)}\right]; \quad (11)$$

unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

$$\dot{m}_{GHG\,\text{released BS}}\left(\text{kg a}^{-1}\right) = \dot{m}_{CO_2\,\text{released biogas combustion or upgrading BS}}\left(\text{kg a}^{-1}\right) + \sum_{\text{unit}} \dot{m}_{CO_2\ \text{leaked BS}_{\text{unit}}}\left(\text{kg a}^{-1}\right) + \sum_{\text{unit}} \left(\dot{m}_{CH_4\ \text{leaked BS}_{\text{unit}}}\left(\text{kg a}^{-1}\right) \times GWP_{CH_4}\right); \quad (12)$$

unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

2.2 Carbon capture technologies and scenarios

Retrofitting WRRF biogas utilization systems with CCS systems would enable the generation of negative GHG emissions by capturing the carbon dioxide in the biogas, the carbon dioxide in combustion off-gases, or the carbon dioxide separated during biogas upgrading to RNG. A survey of the CCS technologies that could be applied, and how their effectiveness can be estimated, is described in this section. We note that the generation of negative emissions hinges on permanent carbon storage. The logistics and costs of carbon dioxide transport and permanent storage are out of the scope of this work.

2.2.1 Technologies for carbon capture from biogas production and valorization. There exist a variety of carbon capture processes from gas streams, including wet scrubbing,[25] pressure and temperature swing adsorption,[26,27] amine absorption,[26,28] precipitation as carbonates,[29] and membrane separation.[30] Carbon dioxide capture and sequestration efficiencies range from 81 to 98%. Another alternative that has been explored is carbon capture within the digester unit by using wollastonite ($CaSiO_3$), achieving sequestration efficiencies of 26–48%.[31,32] Table 3 describes three theoretical carbon capture scenarios for WRRFs, lists the CCS technologies (j) proposed for each scenario, and their range recovery efficiencies uncertainties, denoted as $\eta_{CO_2,\,j}$. The carbon capture estimates for each scenario are described in the proceeding subsections.

2.2.1.1 Scenario 1: CCS from biogas combustion exhaust gases or carbon dioxide from RNG production. Scenario 1 (S1) was defined as the implementation of technologically mature CCS processes for capturing the carbon dioxide from biogas combustion exhaust gases, and/or the carbon dioxide separated from biogas during RNG upgrading. A set of the most commonly deployed CCS processes was assessed, including pressure and temperature swing adsorption, amine adsorption, ammonia wet scrubbing, sorption in carbonate solutions, and membrane-based systems.[26]

The annual mass flow of carbon dioxide captured, $\dot{m}_{CO_2\ \text{captured S1 pc}}$, was estimated based on the mass of biosolids processed through eqn (13). The subscript "S1" denotes Scenario 1 and "pc" denotes the set of post-combustion CCS technologies. The total annual mass flow of GHG emissions as CO_{2eq} after CCS process implementation, $\dot{m}_{GHG\ \text{released S1 pc}}$, was estimated with eqn (14). The uncertainty in the recovery efficiency of each process pc, denoted as $\eta_{CO_2\ pc}$, was addressed by assessing the minimum and maximum efficiency values reported in literature, as shown in Table 3.

$$\dot{m}_{CO_2\ \text{captured S1 pc}}\left(\text{kg a}^{-1}\right) = \dot{m}_{CO_2\ \text{released biogas combustion or upgrading BS}}\left(\text{kg a}^{-1}\right) \times \eta_{CO_2\ pc}; \quad (13)$$

∀ pc ∈ {temperature/pressure swing adsorption, amines absorption, amine wet scrubbing, sorption in carbonates, membrane-based systems}

$$\dot{m}_{GHG\ \text{released S1 pc}}\left(\text{kg a}^{-1}\right) = \dot{m}_{CO_2\ \text{released biogas combustion or upgrading BS}}\left(\text{kg a}^{-1}\right) - \dot{m}_{CO_2\ \text{captured S1 pc}}\left(\text{kg a}^{-1}\right)) + \sum_{\text{unit}} \dot{m}_{CO_2\ \text{leaked BS}_{\text{unit}}}\left(\text{kg a}^{-1}\right) + GWP_{CH_4} \times \sum_{\text{unit}} \left(\dot{m}_{CH_4\ \text{leaked BS}_{\text{unit}}}\left(\text{kg a}^{-1}\right)\right); \quad (14)$$

∀ pc ∈ {temperature/pressure swing adsorption, amines absorption, amine wet scrubbing, sorption in carbonates, membrane-based systems};unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

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Table 3 Scenarios assessed and capture efficiency (η_{CO_2}) of the carbon capture processes selected. Data on carbon dioxide capture efficiency and operational energy requirements for Scenario 3 are combinations of Scenarios 1 and 2

Scenario	Process	CCS technology	$\eta_{CO_2\ min}$ (% relative to capture unit inlet)	$\eta_{CO_2\ max}$ (% relative to capture unit inlet)	Energy requirements (kJ kg^{-1} CO_2 captured)	References
Scenario 1	Post-combustion/ biogas upgrading carbon sequestration	Temperature/pressure swing adsorption	81	97	3000	26, 27, and 33
		Amines absorption	83	98	3800	25, 26, 28, and 33
		Ammonia wet scrubbing	90	90	1147	29 and 33
		Sorption in carbonates	80	90	1179	34 and 35
		Membrane-based systems	83	91	8000	30 and 33
Scenario 2	In-digester precipitation	In-digester carbon mineralization	26	48	58	31, 32, and 36
Scenario 3	In-digester precipitation + post-combustion/biogas upgrading carbon sequestration	In-digester carbon mineralization + temperature/pressure swing adsorption	26 (in-digester); 81 (post-combustion)	48 (in-digester); 97 (post-combustion)	58 (in-digester); 3000 (post-combustion)	—
		In-digester carbon mineralization + amines absorption	26 (in-digester); 83 (post-combustion)	48 (in-digester); 98 (post-combustion)	58 (in-digester); 3800 (post-combustion)	—
		In-digester carbon mineralization + ammonia wet scrubbing	26 (in-digester); 90 (post-combustion)	48 (in-digester); 90 (post-combustion)	58 (in-digester); 1147 (post-combustion)	—
		In-digester carbon mineralization + sorption in carbonates	26 (in-digester); 80 (post-combustion)	48 (in-digester); 90 (post-combustion)	58 (in-digester); 1179 (post-combustion)	—
		In-digester carbon mineralization + membrane separation	26 (in-digester); 83 (post-combustion)	48 (in-digester); 91 (post-combustion)	58 (in-digester); 8000 (post-combustion)	—

2.2.1.2 Scenario 2: CCS by in-digester carbon precipitation. Scenario 2 (S2) is based on carbon mineralization during anaerobic digestion that captures and stores carbon dioxide as precipitated calcium carbonate using a sources of alkalinity and calcium, such as wollastonite.[32] As the calcium carbonate precipitates in the digestate, he permanent storage of this captured carbon dioxide will depend on the conditions and end use of the digestate. Carbon mineralization during anaerobic digestion is a technology under development, with a TRL of 4–5 [31] that captures only the carbon dioxide in biogas. The motivations for this process include increasing the biogas methane concentration and its energy density, and decreasing the biogas volume, which would decrease downstream gas system management capital and operating costs. There exists some controversy about biogas production increase[32] or decrease[31] with in-digester CCS. In this work, no change in biogas production is assumed for the in-digester carbon mineralization process.

Estimation of methane-concentrated biogas leaks and the biogas combustion emissions for Scenario 2 were modified from the baseline biogas generation scenario, as described in eqn (15)–(22), where the subscript S2 denotes "Scenario 2". It was assumed that the fraction of biogas relative to the biogas produced at each stage, $\kappa_{biogas\ unit}$, as calculated by eqn (20), remains constant. The fraction of captured carbon dioxide and the resulting GHG emission estimates were calculated with eqn (20)–(24), where the recovery efficiency is denoted as $\eta_{CO_2\ in\text{-}digester}$. The uncertainty in the recovery efficiency was addressed by evaluating the minimum and maximum efficiency values reported in the literature, which are collected in Table 3.

$$\dot{m}_{CO_2\ captured\ S2}\left(kg\ a^{-1}\right) = \dot{m}_{CO_2\ produced\ BS}\left(kg\ a^{-1}\right) \times \eta_{CO_2\ in\text{-}digester} \quad (15)$$

$$\dot{m}_{CH_4\ produced\ S2}\left(kg\ a^{-1}\right) = \dot{m}_{CH_4\ produced\ BS}\left(kg\ a^{-1}\right) \quad (16)$$

$$\dot{m}_{CO_2\ produced\ S2}\left(kg\ a^{-1}\right) = \dot{m}_{CO_2\ produced\ BS}\left(kg\ a^{-1}\right) - \dot{m}_{CO_2\ captured\ S2}\left(kg\ a^{-1}\right) \quad (17)$$

$$\dot{m}_{biogas\ produced\ S2}\left(kg\ a^{-1}\right) = \left(\frac{\dot{m}_{CH_4\ produced\ S2}\left(kg\ a^{-1}\right)}{MW_{CH_4}\left(kg\ kmol^{-1}\right)} + \frac{\dot{m}_{CO_2\ produced\ S2}\left(kg\ a^{-1}\right)}{MW_{CO_2}\left(kg\ kmol^{-1}\right)}\right) \times MW_{biogas}\left(kg\ kmol^{-1}\right) \quad (18)$$

$$x_{i\ S2} = \frac{\frac{\dot{m}_{i\ produced\ S2}\left(kg\ a^{-1}\right)}{MW_i\left(kg\ kmol^{-1}\right)}}{\sum_i \frac{\dot{m}_{i\ produced\ S2}\left(kg\ a^{-1}\right)}{MW_i\left(kg\ kmol^{-1}\right)}} \quad (19)$$

$\forall\ i \in \{CH_4, CO_2\}$

$$\kappa_{biogas\ unit}(\%) = \frac{\dot{m}_{biogas\ leaked\ BS_{unit}}\left(kg\ a^{-1}\right)}{\dot{m}_{biogas\ produced\ BS}\left(kg\ a^{-1}\right)} \times 100; \quad (20)$$

$\forall$ unit $\in$ {digester and feeding system, digestate storage, biogas combustion, RNG production}

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$$\dot{m}_{\mathrm{CH_4\ leaked\ S2\ unit}}\left(\mathrm{kg\ a^{-1}}\right) = \frac{\dot{m}_{\mathrm{biogas\ produced\ S2}}\left(\mathrm{kg\ a^{-1}}\right)\cdot\kappa_{\mathrm{biogas\ unit}}(\%)}{\mathrm{MW_{biogas}}\left(\mathrm{kg\ kmol^{-1}}\right)} \times x_{\mathrm{CH_4\ S2}} \times \mathrm{MW_{CH_4}}\left(\mathrm{kg\ kmol^{-1}}\right); \quad (21)$$

∀ unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

$$\dot{m}_{\mathrm{CO_2\ leaked\ S2\ unit}}\left(\mathrm{kg\ a^{-1}}\right) = \frac{\dot{m}_{\mathrm{biogas\ produced\ S2}}\left(\mathrm{kg\ a^{-1}}\right)\cdot\kappa_{\mathrm{biogas\ unit}}(\%)}{\mathrm{MW_{biogas}}\left(\mathrm{kg\ kmol^{-1}}\right)} \times x_{\mathrm{CO_2\ S2}} \times \mathrm{MW_{CO_2}}\left(\mathrm{kg\ kmol^{-1}}\right); \quad (22)$$

∀ unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

$$\dot{m}_{\mathrm{CO_2\ released\ biogas\ combustion\ or\ upgrading\ S2}}\left(\mathrm{kg\ a^{-1}}\right) = \dot{m}_{\mathrm{CO_2\ produced\ S2}}\left(\mathrm{kg\ a^{-1}}\right) - \sum_{\mathrm{unit}}\dot{m}_{\mathrm{CO_2\ leaked\ S2\ unit}}\left(\mathrm{kg\ a^{-1}}\right) + \left[\dot{m}_{\mathrm{CH_4\ produced\ S2}}\left(\mathrm{kg\ a^{-1}}\right) - \sum_{\mathrm{unit}}\dot{m}_{\mathrm{CH_4\ leaked\ S2\ unit}}\left(\mathrm{kg\ a^{-1}}\right)\right] \times \frac{\mathrm{MW_{CO_2}}\left(\mathrm{kg\ kmol^{-1}}\right)}{\mathrm{MW_{CH_4}}\left(\mathrm{kg\ kmol^{-1}}\right)}; \quad (23)$$

unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

$$\dot{m}_{\mathrm{GHG\ released\ S2}}\left(\mathrm{kg\ a^{-1}}\right) = \dot{m}_{\mathrm{CO_2\ released\ biogas\ combustion\ or\ upgrading\ S2}}\left(\mathrm{kg\ a^{-1}}\right) + \sum_{\mathrm{unit}}\dot{m}_{\mathrm{CO_2\ leaked\ S2\ unit}}\left(\mathrm{kg\ a^{-1}}\right) + \mathrm{GWP_{CH_4}} \times \sum_{\mathrm{unit}}\left(\dot{m}_{\mathrm{CH_4\ leaked\ S2\ unit}}\left(\mathrm{kg\ a^{-1}}\right)\right); \quad (24)$$

unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

2.2.1.3 Scenario 3: Integrating CCS by in-digester carbon mineralization precipitation and from post-combustion or RNG production. Scenario 3 integrates both in-digester carbon mineralization and CCS systems for capturing carbon dioxide from on-site post-combustion or biogas upgrading streams. This scenario evaluates the synergetic effect of capturing carbon dioxide during biogas generation by in-digester carbon mineralization, which may result in lower overall biogas leak rates, and the higher carbon capture efficiency anticipated from post-combustion or biogas upgrading from methane-rich streams. The mass flows of carbon dioxide captured and GHG emissions estimated for Scenario 3 are described in eqn (25)–(27).

$$\dot{m}_{\mathrm{CO_2\ captured\ biogas\ combustion\ or\ upgrading\ S3\ pc}}\left(\mathrm{kg\ a^{-1}}\right) = \dot{m}_{\mathrm{CO_2\ released\ biogas\ combustion\ or\ upgrading\ S2}}\left(\mathrm{kg\ a^{-1}}\right) \times \eta_{\mathrm{CO_2\ pc}}; \quad (25)$$

∀ pc ∈ {temperature/pressure swing adsorption, amines absorption, amine wet scrubbing, sorption in carbonates, membrane-based systems}

$$\dot{m}_{\mathrm{CO_2\ captured\ S3\ pc}}\left(\mathrm{kg\ a^{-1}}\right) = \dot{m}_{\mathrm{CO_2\ captured\ biogascombustion\ or\ upgrading\ S3\ pc}}\left(\mathrm{kg\ a^{-1}}\right) + \dot{m}_{\mathrm{CO_2\ captured\ S2}}\left(\mathrm{kg\ a^{-1}}\right) \quad (26)$$

∀ pc ∈ {temperature/pressure swing adsorption, amines absorption, amine wet scrubbing, sorption in carbonates, membrane-based systems}

$$\dot{m}_{\mathrm{GHG\ released\ S3\ pc}}\left(\mathrm{kg\ a^{-1}}\right) = \left(\dot{m}_{\mathrm{CO_2\ released\ biogas\ combustion\ or\ upgrading\ S2}} - \dot{m}_{\mathrm{CO_2\ captured\ S3\ pc}}\right)\left(\mathrm{kg\ a^{-1}}\right) + \sum_{\mathrm{unit}}\dot{m}_{\mathrm{CO_2\ leaked\ S2\ unit}}\left(\mathrm{kg\ a^{-1}}\right) + \mathrm{GWP_{CH_4}} \times \sum_{\mathrm{unit}}\left(\dot{m}_{\mathrm{CH_4\ leaked\ S2\ unit}}\left(\mathrm{kg\ a^{-1}}\right)\right); \quad (27)$$

∀ pc ∈ {temperature/pressure swing adsorption, amines absorption, amine wet scrubbing, sorption in carbonates, membrane-based systems};
unit ∈ {digester and feeding system, digestate storage, biogas combustion, RNG production}

2.2.2 Carbon emissions from carbon capture technologies. Carbon dioxide capture through the different evaluated processes requires energy, as shown in Table 3. To account for the GHG emissions generated by the energy demands of the carbon capture processes, we assumed that the energy is supplied through the electric grid of each province. The provincial electric grids in Canada carry electric power generated by a mixture of power generation technologies. Across Canada, the provincial average GHG emissions for electric power vary widely. Provinces with predominantly hydroelectric power stations, such as Quebec, Manitoba and British Columbia, generate less GHG per kWh than other provinces where a significant fraction of electric power is generated by fossil fuel combustion, such as Nova Scotia and Saskatchewan. In order to account for the additional carbon emissions derived from electricity transport from production to consumption sites, the carbon intensity per unit of energy consumed was considered.[37] The estimation of carbon emissions from carbon capture technologies is described in eqn (28), where E_{CC} denotes the energy requirements of the carbon capture (CC) system, and $\mathrm{CI_{province}}$ denotes the carbon intensity of each province, which are listed in Table 4.

$$\dot{m}_{\mathrm{CO_2\ emissions\ CC}}\left(\mathrm{kg\ a^{-1}}\right) = \dot{m}_{\mathrm{CO_2\ captured\ CC}}\left(\mathrm{kg\ a^{-1}}\right) \times E_{\mathrm{CC}}\left(\mathrm{kJ\ kg_{\ CO_2\ captured}^{-1}}\right) \times \mathrm{CI_{province}}\left(\mathrm{kg\ CO_{2\ eq}\ kJ^{-1}}\right) \quad (28)$$

∀ CC ∈ {temperature/pressure swing adsorption, amines absorption, amine wet scrubbing, sorption in carbonates, membrane-based systems, in-digester mineralization}
province ∈ {Alberta, British Columbia, Manitoba, Newfoundland and Labrador, Nova Scotia, Ontario, Prince Edward Island, Quebec, Saskatchewan, New Brunswick}

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Table 4 Carbon intensity for electricity consumption[37]

Province	Intensity ($g\ CO_{2\ eq}$ per kWh_e consumed)
Alberta	474.0
British Columbia	15.6
Manitoba	1.4
Newfoundland and Labrador	17.9
Nova Scotia	690.0
Ontario	48.5
Prince Edward Island	336.0
Quebec	1.5
Saskatchewan	655.0
New Brunswick	336.0

2.3 Estimation of negative carbon emissions and GHG offset potential for the waste management sector

The capture of one kilogram of biogenic carbon was assumed to be equivalent to the generation of one kilogram of negative carbon emissions. As the carbon in raw biogas contains 98.4% biogenic carbon,[8] it is assumed that the capture of one kilogram of carbon from biogas was equivalent to the generation of 0.984 kilograms of carbon negative emissions.

GHG emissions from the Canadian waste management sector were collected from the Canada's Official Greenhouse Gas Inventory[37] for 2021, the most recent year at the time that the study was performed. The potential negative emissions generated by biogenic carbon capture were assessed for all the provincial waste management sector categories reported by the Canada's Official Greenhouse Gas Inventory.

2.4 Computational framework

Fig. 2 shows the framework utilized to estimate the GHG emitted by the production and utilization of biogas in the Canadian WRRFs. The Monte Carlo method was used to address the uncertainties associated with sewage sludge production rates reported in Table 1, organic matter degradation, biogas composition, and biogas leaks. While normal distribution for biogas composition has been reported,[3] we selected a triangular distribution for the remaining variables due to the lack of information regarding their distribution functions. The values reported for the minimum and maximum carbon capture scenarios denote the upper and lower limit of the 95% confidence interval.

The number of Monte Carlo iterations (n) were estimated through eqn (29), which is derived from the Central Limit Theorem.[38] The variable utilized to estimate (n) was the total GHG emissions, assuming a confidence interval of 95% ($z_{\alpha/2}$ equal to 1.96) and an error (ε) if 1%. σ_{sample} and μ_{sample} denote the average and standard deviation of a sample of the variable obtained through an prospective run of the Monte Carlo model with 100 iterations. σ_{sample} and μ_{sample} were used as estimators of the real average and standard deviation of the dependent variables (*i.e.*, the total GHG emissions), which are unknown. n was estimated for all assessed WRRFs, using the largest n among the values obtained as the number of Monte Carlo iterations used by the framework for estimating the GHG emissions of all studied WRRFs.

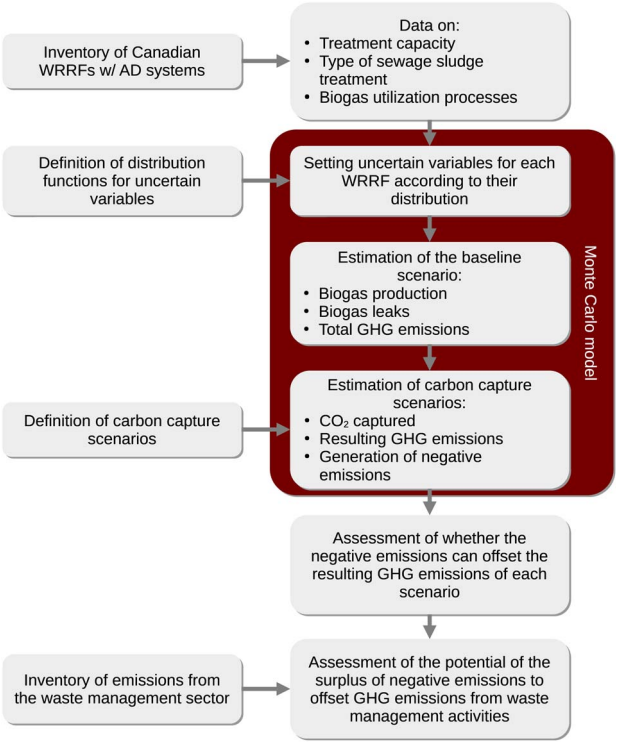

Fig. 2 Computational framework used to estimate biogas production, carbon dioxide and GHG emissions, carbon captured and offset potential for the biogas production and utilization systems at the Canadian WRRFs equipped with AD systems.

$$n = \left(z_{\alpha/2} \frac{100 \times \sigma_{sample}}{\varepsilon \mu_{sample}} \right)^2 \quad (29)$$

3 Results and discussion

The estimated carbon emission inventory from biogas production and use in Canadian WRRFs with biogas production will be described, followed by the effects of the three CCS scenarios described in Section 2.2.

3.1 Inventory of carbon emissions from biogas production and use at Canada's municipal WRRFs

3.1.1 Mapping biogas production and use in Canada's municipal WRRFs. Fig. 3a shows the wide implementation of AD systems for sewage sludge treatment across Canadian WRRFs. WRRFs equipped with AD systems account for a large share of the treated wastewater, although their ratio in terms of number of facilities is considerably lower, indicating that biogas production is predominantly carried out in large-scale WRRFs. A total of 93 AD systems installed in municipal WRRFs were identified.

Fig. 3b shows the spatial distribution and size of biogas production systems installed at WRRFs, as well as the biogas

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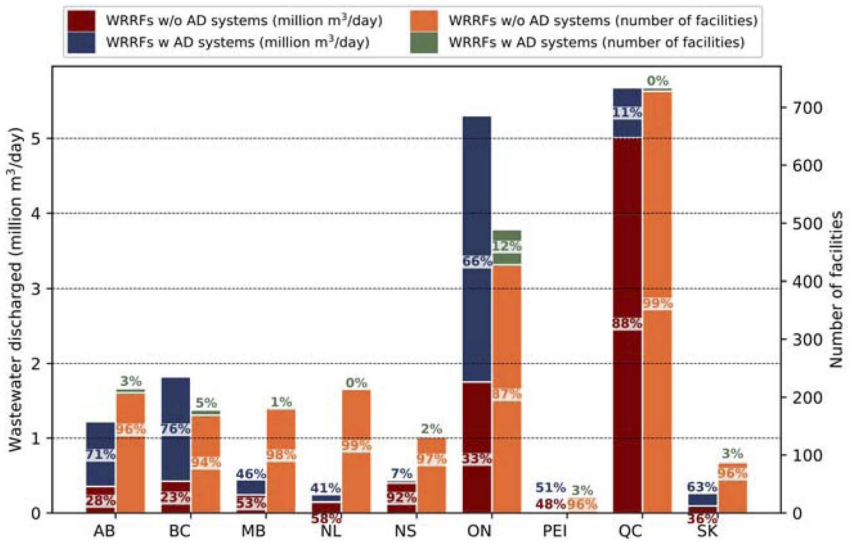
use at each WRRF, including biogas flaring and the production of heat, electricity, and RNG production. AD systems are implemented in medium (discharging 15 500–31 000 m^3/day) and large (discharging >31 000 m^3/day) WRRFs.[39] These facilities are predominately situated along the southern border, where most of the largest urban centers are located. The Quebec City–Windsor corridor is a region located to the north of the St. Lawrence River and Lake Erie between Quebec City, Quebec, and Windsor, Ontario. This corridor includes the cities of Montreal, Ottawa, and Toronto, and is where a high density of WRRFs equipped with AD systems are located. The largest AD facilities are located in the WRRFs that serve most of the large urban centers, with the notable exception of the WWTP of the City of Montreal. Montreal is the largest city in Quebec, and is situated on a river island. Due to the limited land available, the Montreal WWTP process includes physical and chemical treatments without AD or other biological treatment, followed by sludge incineration.

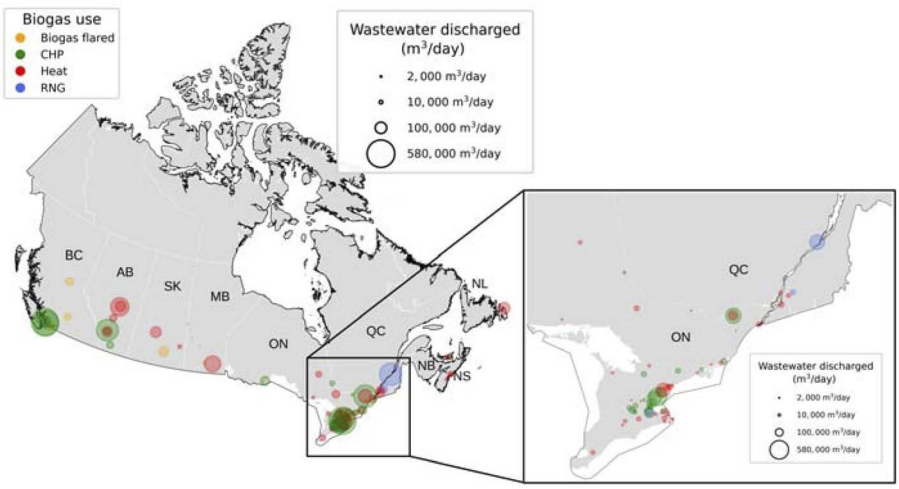
(a) Biogas production within the Canadian wastewater treatment sector.

(b) Locations and sizes of anaerobic digestion (AD) systems at Canadian WRRFs.

3.1.2 Relationships between biogas production rates and uses, and population served. The relationship between the provincial population and provincial biogas production is presented in Fig. 4a. 214 kt/a of biogas are produced at Canadian municipal WRRFs. Assuming a low heating value of 20 MJ kg^{-1}, 4.3 PJ/a of biogas is produced by WRRFs, representing 19.5% of the total annual biogas production in Canada, *i.e.*, 22 PJ.[40] Fig. 4a shows the biogas uses for the Canadian provinces with



(a) Biogas production within the Canadian wastewater treatment sector.



(b) Locations and sizes anaerobic digestion (AD) systems at Canadian WRRFs.

Fig. 3 Share, locations, and size of anaerobic digestion (AD) systems at Canadian WRRFs. New Brunswick (NB) data were not included in the figure because no WRRFs with AD were reported for this province. AB: Alberta, BC: British Columbia, MB: Manitoba, NL: Newfoundland and Labrador, NS: Nova Scotia, ON: Ontario, PEI: Prince Edward Island, QC: Quebec, SK: Saskatchewan.

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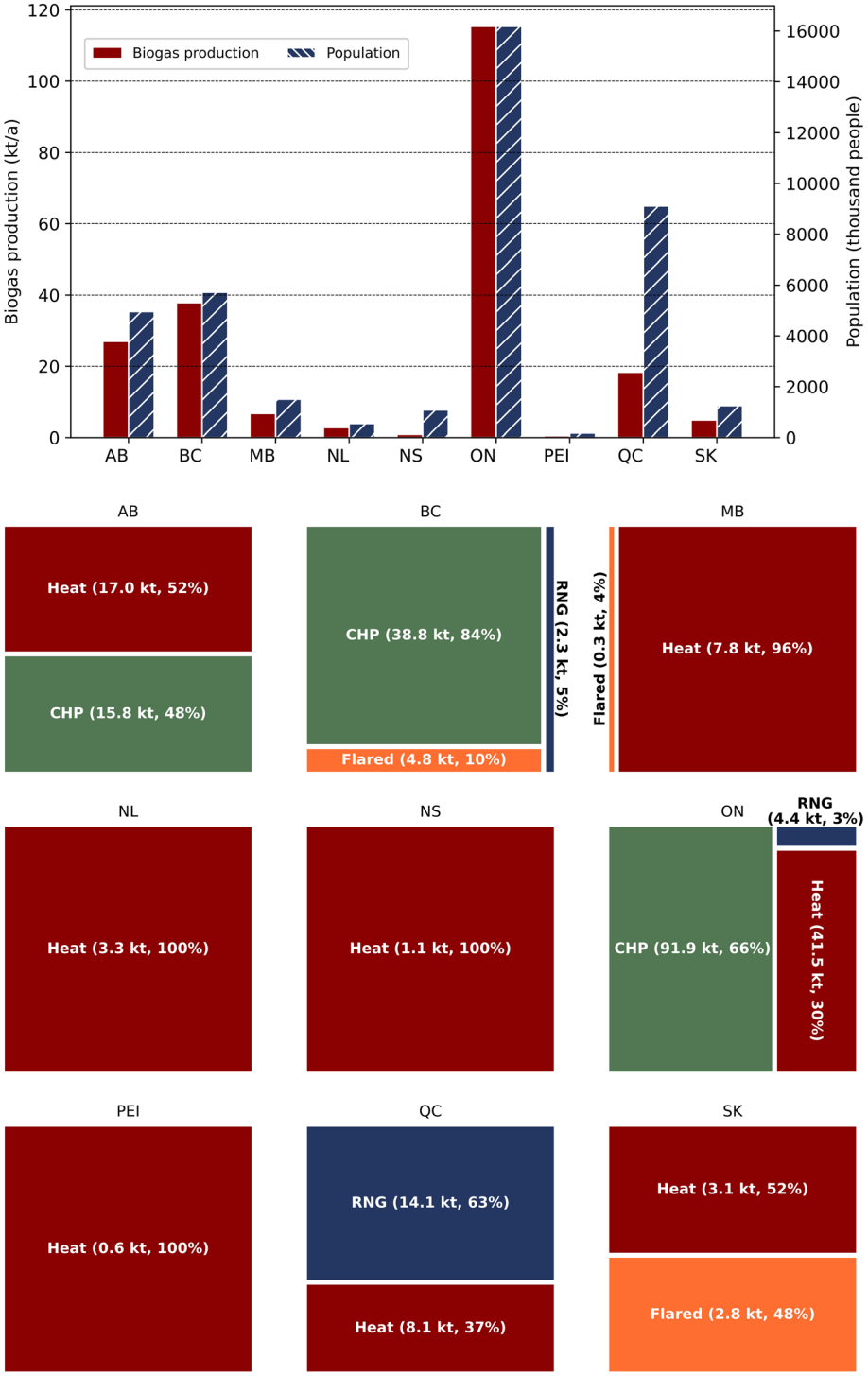

Fig. 4 Population, annual biogas production rates, and biogas applications at Canadian WRRFs per province. New Brunswick (NB) was not included because it did not report any biogas-generating WWTPs. kt denotes kilotonne. AB: Alberta, BC: British Columbia, MB: Manitoba, NL: Newfoundland and Labrador, NS: Nova Scotia, ON: Ontario, PEI: Prince Edward Island, QC: Quebec, SK: Saskatchewan. (a) Uses of biogas at Canadian WRRFs.

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AD in at least one WRRF. It can be observed that for most provinces, there exists a direct relationship between biogasproduction at WRRFs and population. Quebec is an exception as its largest facility, the WRRF of the City of Montreal, is equipped with sludge incineration.

(a) Provincial biogas production rates and populations for Canadian provinces with AD at WRRFs.

The most common biogas use is thermal energy production to meet the WRRF process and AD unit demands, and occasionally for heating buildings. The next most common biogas use is combined heat and power (CHP) units that generate thermal and electrical energy. A small number of biogas upgrading facilities produce RNG at WRRFs in British Columbia, Ontario, and Quebec. Finally, biogas is flared without energy recovery in some WWRF facilities in British Columbia, Manitoba, and Saskatchewan. Flaring reduces the benefits of biogas production, while reducing the GHG emissions by converting higher GWP methane to carbon dioxide before its emission.

3.1.3 GHG emissions from biogas production and utilization at municipal WRRFs. Fig. 5 shows the GHG emissions estimates for each Canadian province and biogas use, as well as provincial totals, while Fig. 6 illustrates the breakdown of GHG emissions at each stage of the biogas production and utilization processes. The two major sources of GHG emissions are the biogas leaks from the digestate production and storage before solid/liquid separation, and the biogas combustion off-gasses during biogas utilization. Similar results are observed for all provinces.

Ontario, the province with largest population, generated the largest Canadian GHG emissions from its AD systems (277.7 ± 1.9 kt $CO_{2\ eq}\ a^{-1}$), followed by British Columbia (92.5 ± 1.2 kt $CO_{2\ eq}\ a^{-1}$) with the third largest population, and Alberta 66.1 ± 0.9 kt $CO_{2\ eq}\ a^{-1}$) with the fourth largest population.

Biogas use and management varies between the provinces. Biogas combustion for heat generation and cogeneration of heat and electricity (CHP) are the main sources of biogas GHG emissions in Ontario and Alberta. In Ontario, GHG emissions for CHP (185.6 ± 3.5 kt $CO_{2\ eq}\ a^{-1}$) were estimated to be more than double that GHG emissions for heat generation (83.9 ± 1.6 kt $CO_{2\ eq}\ a^{-1}$). In Alberta, heat generation (34.3 ± 1.0 $CO_{2\ eq}\ a^{-1}$) and the use of CHP units (31.9 ± 1.5 kt $CO_{2\ eq}\ a^{-1}$) released similar GHG magnitudes. British Columbia did not report any standalone heat generation, while most of the GHG releases were from CHP units (78.5 ± 2.5 kt $CO_{2\ eq}\ a^{-1}$) and biogas flaring without energy recovery (9.7 ± 0.3 kt $CO_{2\ eq}\ a^{-1}$). Two provinces upgrade a small share of their biogas to RNG: British Columbia upgraded 4.7% of its biogas production, while Ontario upgraded 2.9% of its biogas production. Due to the larger biogas production rate in Ontario, the GHG emissions from Ontario biogas upgrading (8.1 ± 0.4 kt $CO_{2\ eq}\ a^{-1}$) were larger than for British Columbia (4.3 ± 0.2 kt $CO_{2\ eq}\ a^{-1}$).

The province of Quebec has a different GHG emission pattern due to two major RNG production projects; one in Quebec City, and the second in Saint-Hyacinthe. As a result, the largest Quebec GHG emission was from RNG production (25.9 ± 1.2 kt $CO_{2\ eq}\ a^{-1}$), followed by heat recovery (16.5 ± 0.5 kt $CO_{2\ eq}\ a^{-1}$).

GHG emissions from other provinces were produced by less complex biogas valorization technologies. Heat recovery by biogas combustion was reported for the provinces of Manitoba (15.8 ± 0.8 kt $CO_{2\ eq}\ a^{-1}$), Newfoundland and Labrador (6.8 ± 0.3 kt $CO_{2\ eq}\ a^{-1}$), Saskatchewan (6.3 ± 0.3 kt $CO_{2\ eq}\ a^{-1}$), Nova Scotia (2.3 ± 0.1 kt $CO_{2\ eq}\ a^{-1}$), and Prince Edward Island (1.3 ± 0.1 kt $CO_{2\ eq}\ a^{-1}$). Biogas flaring was a significant GHG emission in the province of Saskatchewan (5.7 ± 0.3 kt $CO_{2\ eq}\ a^{-1}$), while this process represents a marginal share of GHG emissions in Manitoba (0.6 ± 0.1 kt $CO_{2\ eq}\ a^{-1}$).

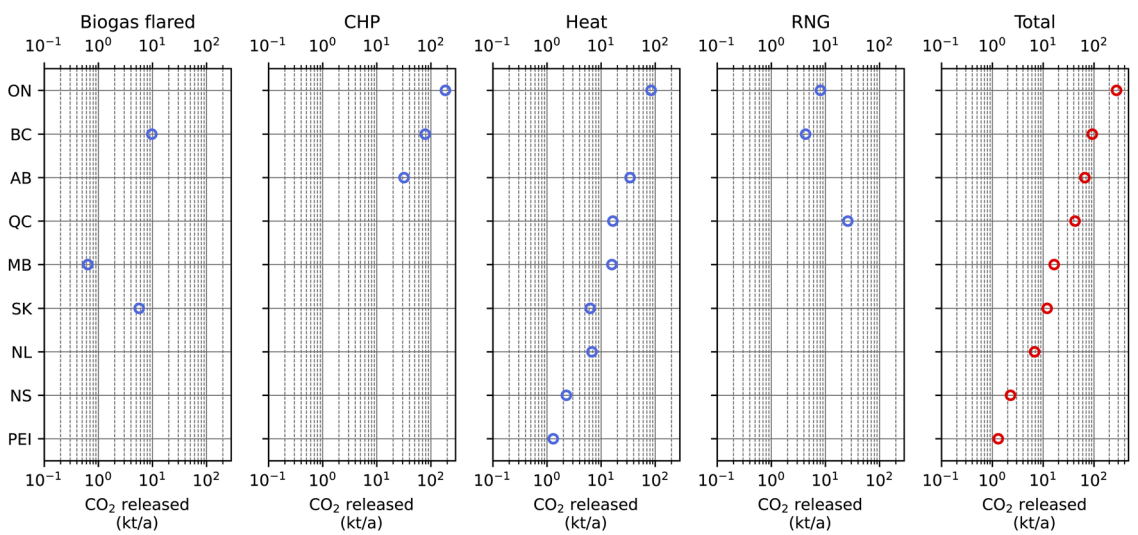

Fig. 5 GHG emissions from biogas production and use at municipal WRRFs per province and type of biogas-use process, and province totals. New Brunswick (NB) data were not included in the figure as no WRRFs with AD were reported for this province. kt denotes kilotonne. AB: Alberta, BC: British Columbia, MB: Manitoba, NL: Newfoundland and Labrador, NS: Nova Scotia, ON: Ontario, PEI: Prince Edward Island, QC: Quebec, SK: Saskatchewan.

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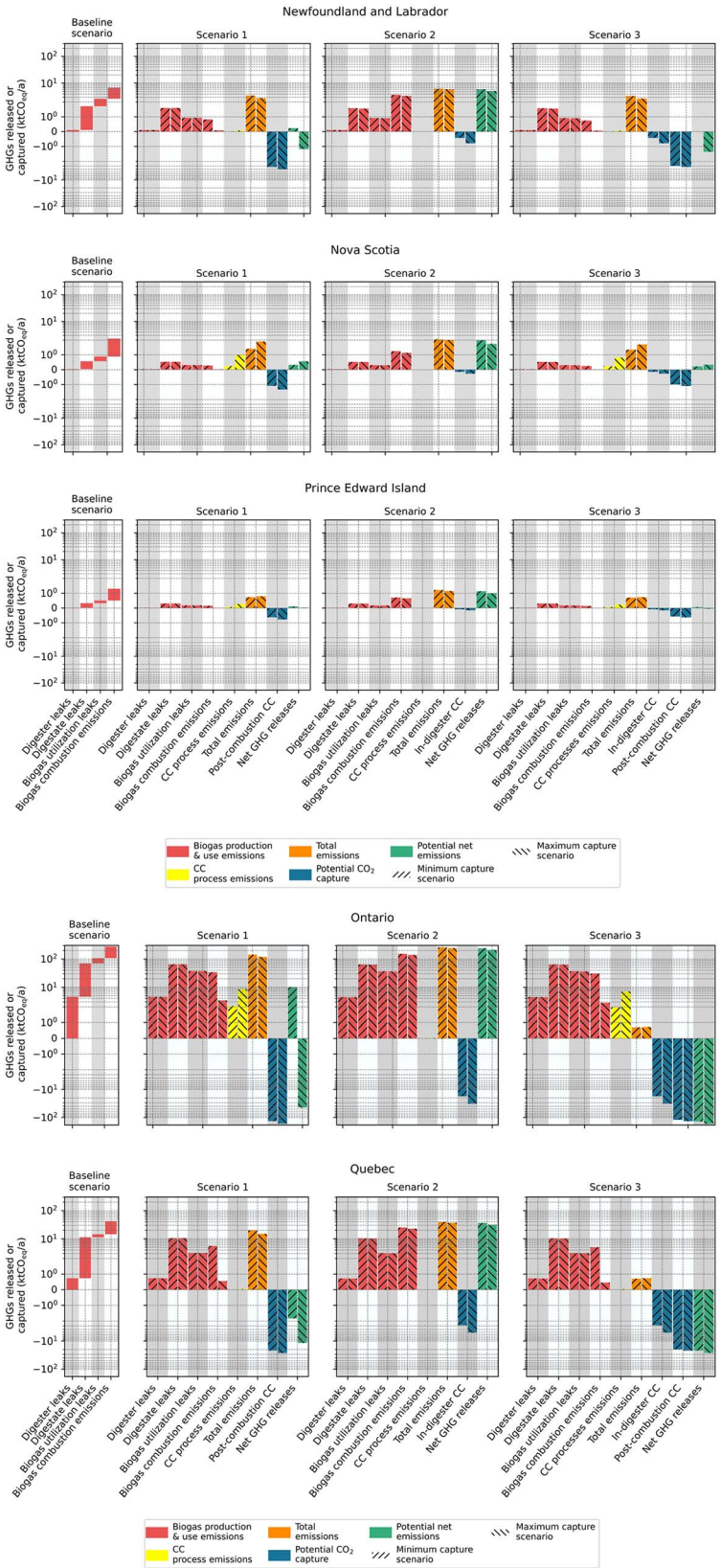

Fig. 6 Detailed carbon dioxide emissions, abatement potential and net emissions from biogas production and use at municipal WRRFs per province. New Brunswick data were not included in the figure because no WRRFs with AD were reported for this province. Scenario 1 shows the implementation of post-combustion/-biogas upgrading CCS systems, Scenario 2 shows CCS by in-digester carbon mineralization, and Scenario 3 shows the combination both of these technologies. kt denotes kilotonne.

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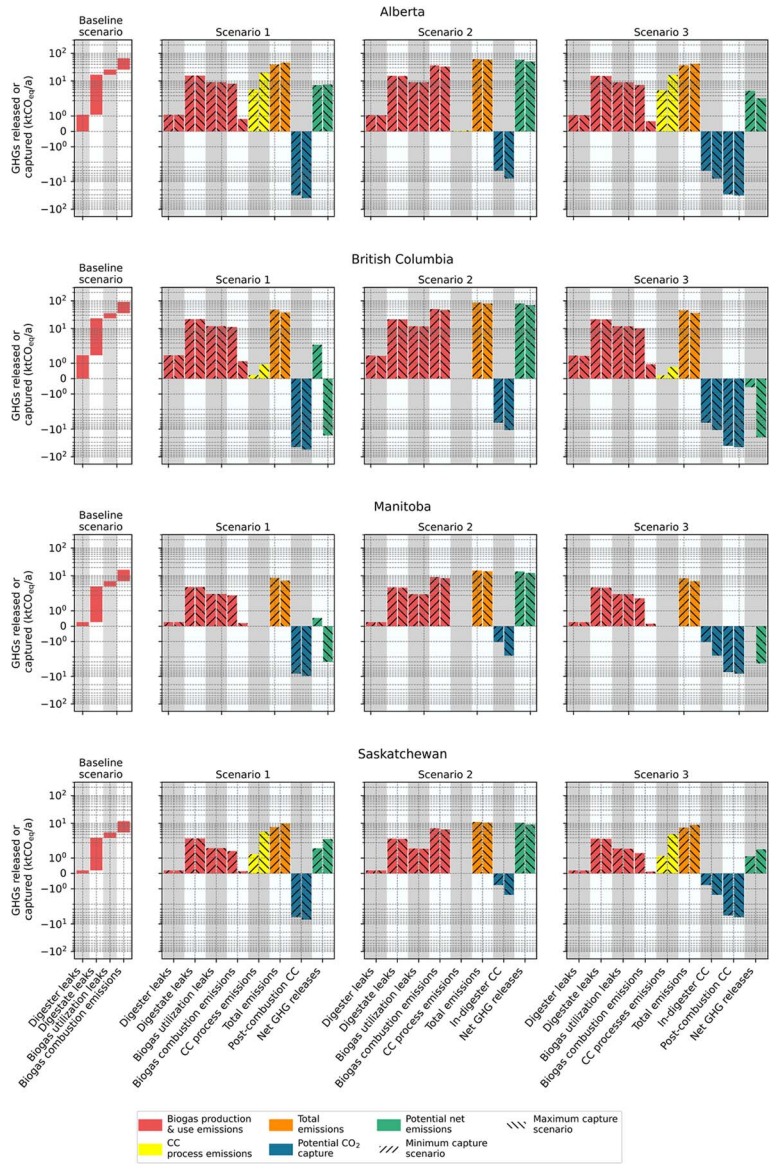

Fig. 6 Fig. 6 (*Contd.*)

3.2 Decarbonizing biogas production and utilization at municipal WRRFs

3.2.1 Carbon dioxide capture estimates. GHG emissions related to biogas generation and use at WRRFs are from uncontrolled leaks and controllable emissions. A significant fraction of WRRF biogas emissions are from leaks at the different stages of biogas production and utilization. The methane component of these leaks increases the GHG emission due to the higher GWP of methane. These GHG emissions from leaks cannot be captured. GHG emissions from carbon dioxide emissions after combustion or by RNG produciton are suitable for carbon capture.

During the last decades, several carbon capture and sequestration technologies have been designed and tested,[29] some of which have reached full operational development.[41] However, WRRF biogas production and utilization processes are usually not equipped with carbon capture systems,[15] When considering all GHG emissions from WWRFs, methane is the gas with the highest GWP, while carbon dioxide from biogas combustion off-gases does not contribute to the net WRRF fossil GHG emission inventory. Therefore, the implementation of carbon capture processes in WRRF biogas production and utilization has the potential to mitigate the GHG emissions due to methane leaks, leading to a carbon-neutral process. Moreover, if the GHG emission reduction from captured biogenic carbon dioxide exceeds the GHG emissions due to methane leaks, this credit could offset GHG emissions from other sectors.

Fig. 6 shows the estimated GHG emissions for biogas production and utilization, GHG emissions from the carbon

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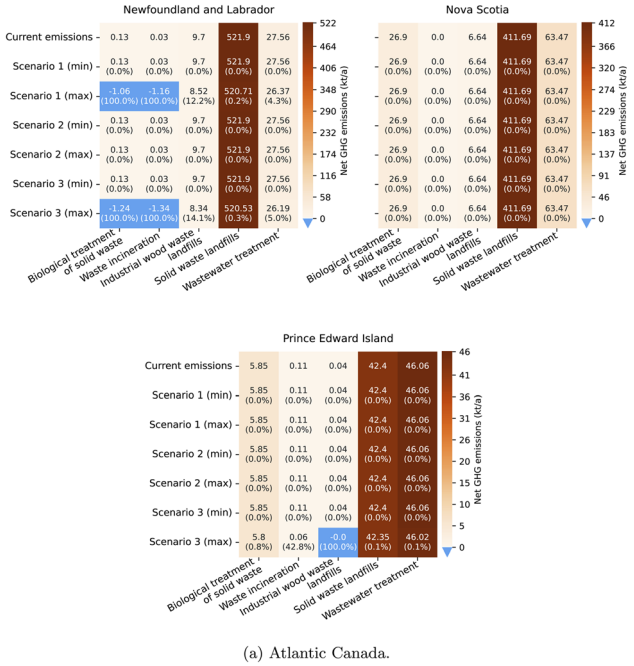
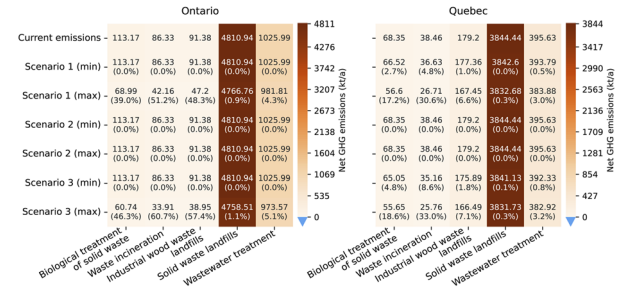
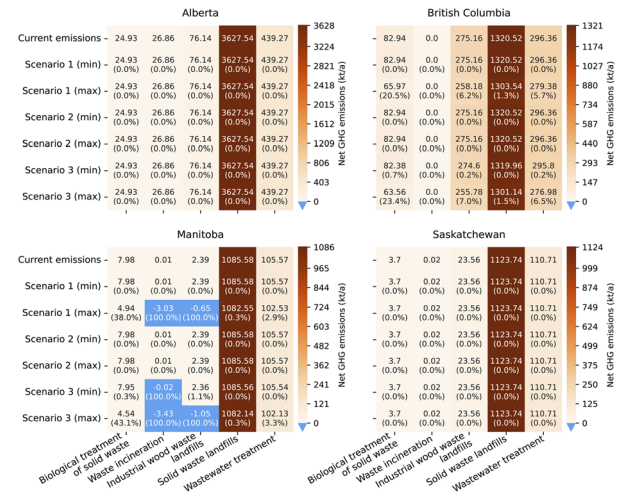


(a) Atlantic Canada.



(b) Eastern Canada.



(c) Western Canada.

Fig. 7 Current GHG emissions from the waste management sector and net emissions after being offset by the surplus of negative emissions obtained from CCS in biogas production and utilization in Canadian WRRFs. The numbers between parenthesis report the share of emissions offset. Blue squares denote those scenarios where the emissions generated by a specific waste management activity are completely offset. New Brunswick (NB) data were not included in the figure because no WRRFs with AD were reported for this province. kt denotes kilotonne.

capture processes, carbon dioxide capture potential, and net GHG emissions after adding CCS processes to the existing WRRF biogas utilization systems for each of nine Canadian provinces. The provincial results are grouped into three geographical areas: Atlantic Canada (Fig. 6a), Eastern Canada (Fig. 6a), and Western Canada (Fig. 6a). The variations in CCS process carbon dioxide capture efficiencies are presented as minimum (left bar, paired by colour) and maximum (right bar, paired by colour) carbon dioxide capture efficiencies for each scenario.

The potential of installing carbon capture systems after biogas combustion or upgrading (Scenario 1) is estimated on the capture of 4.9–6.1 kt $CO_{2\ eq}\ a^{-1}$ in the Atlantic provinces, 155.7–190.7 kt $CO_{2\ eq}\ a^{-1}$ in Eastern Canada, and 90.1–110.4 kt $CO_{2\ eq}\ a^{-1}$ in Western Canada.

In-digester CCS (Scenario 2) estimates captured less $CO_{2\ eq}$ than Scenario 1. This is due to the lower in-digester capture efficiencies compared to the traditional CCS systems, and does not include carbon capture after biogas combustion. Implementing in-digester CCS was estimated to capture 0.7–1.2 kt $CO_{2\ eq}\ a^{-1}$ in the Atlantic provinces, 20.5–37.9 kt $CO_{2\ eq}\ a^{-1}$ in Eastern Canada, and 11.9–21.9 kt $CO_{2\ eq}\ a^{-1}$ in the Western provinces.

Combination of the partial capture of carbon dioxide in raw biogas through in-digester CCS, emitted carbon dioxide, and carbon dioxide produced by methane combustion after biogas utilization (Scenario 3) resulted in the highest estimated GHG emission reduction. Integration of both CCS systems was estimated to reduce the GHG emissions by 5.2–6.2 kt $CO_{2\ eq}\ a^{-1}$ in the Atlantic provinces, 163.4–195.1 kt $CO_{2\ eq}\ a^{-1}$ in the Eastern provinces, and 94.4–112.8 kt $CO_{2\ eq}\ a^{-1}$ in the Western provinces.

3.2.2 Net GHG emissions accounting. When captured, 98% of the carbon dioxide generated directly by AD and indirectly by the combustion of biogas methane was considered to be a negative GHG emission, as a result of the large fraction of biogenic carbon in sewage sludge (see Section 2.3). These negative GHG carbon dioxide emissions generated by implementing CCS systems in WRRFs could offset the GHG emissions from methane leaks in biogas production and utilization. However, the operation of carbon capture systems may generate additional carbon emissions due to their energy requirements, see Table 3, that must be also accounted to quantify the resulting net GHG emissions after implementing carbon capture systems. The net GHG emission estimates are illustrated in Fig. 6 (green bars).

Implementation of carbon dioxide capture systems after biogas combustion or upgrading (Scenario 1) with minimum capture efficiency estimates reduces the carbon dioxide emissions from biogas production and utilization processes to nearly carbon neutral in several provinces. The resulting GHG emissions were 10.1 kt $CO_{2\ eq}\ a^{-1}$ in Ontario, 7.1 kt $CO_{2\ eq}\ a^{-1}$ in Alberta, and 2.6 kt $CO_{2\ eq}\ a^{-1}$ in British Columbia. When the maximum efficiencies of CCS systems after biogas combustion or upgrading were used, some provinces showed net negative GHG emissions, including Newfoundland and Labrador (−1.2 kt $CO_{2\ eq}\ a^{-1}$), Ontario (−44.2 kt $CO_{2\ eq}\ a^{-1}$), Quebec (−11.8 kt

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$CO_{2\ eq}\ a^{-1}$), British Columbia (−17.0 kt $CO_{2\ eq}\ a^{-1}$), and Manitoba (−3.0 kt $CO_{2\ eq}\ a^{-1}$). Regional emissions for this scenario were estimated to be −0.6 kt $CO_{2\ eq}\ a^{-1}$ in the Atlantic provinces, −55.9 kt $CO_{2\ eq}\ a^{-1}$ in the Eastern provinces, and −9.8 kt $CO_{2\ eq}\ a^{-1}$ in the Western provinces. It was observed that the emissions from carbon capture systems are significant in those provinces with high carbon intensities for electricity production, resulting in disadvantageous scenarios for achieving net-zero biogas production processes by offsetting the GHG emissions from methane leaks.

In-digester CCS estimates (Scenario 2) with the negative AD carbon credit do not generate net negative GHG emissions for any province. Furthermore, in-digester carbon mineralization is a process with low energy requirements, and as such, the impact carbon emission from the carbon capture process is more limited than in Scenario 1.

Combining in-digester carbon mineralization with biogas post-combustion or upgrading CCS systems (Scenario 3) offset the total GHG emissions from biogas production and utilization in most provinces. Post-combustion carbon capture systems, which are efficient but energy intensive, are required to capture less carbon dioxide since a fraction is captured upstream by the use of in-digester mineralization, which requires less energy. As a result, the carbon emissions from the carbon capture process were reduced with respect to Scenario 1, and the overall carbon capture performance was improved. However, in those provinces with very high carbon intensities for electricity consumption, no negative emissions were achieved, namely Nova Scotia, Alberta, and Saskatchewan. The regional emissions in the Atlantic provinces were estimated to be range from 0.3 to −1.1 kt $CO_{2\ eq}\ a^{-1}$, in the Eastern provinces the estimates summed up to −159.7 to −193.1 kt $CO_{2\ eq}\ a^{-1}$, while the emissions from the Western provinces were estimated to be 5.0 to −18.8 kt $CO_{2\ eq}\ a^{-1}$.

In addition to the generation of negative emission by capturing biogenic carbon dioxide, it must be noted that the use of biogas also contributed to the mitigation of net GHG emissions by replacing fossil fuels and their associated carbon dioxide emissions. This two-fold contribution of biogas to the reduction of GHG emissions has been explored for Canada, estimating that up to 80 million tonnes of carbon dioxide could be removed.[42] Other assessments of the potential use of negative emissions generated by capturing biogenic carbon from biogas have been conducted for Germany[42] and Poland,[43] emphasizing also the crucial role of carbon intensity of the electricity consumed by the carbon capture processes to obtain negative carbon emissions. Furthermore, the use of negative emissions from biogas production and use has been explored to achieve net-zero processes for the production of different materials, such as methane, ethanol, and electricity.[44,45]

3.3 Application of WRRF GHG emissions offsets to other waste management sector processes

WRRF GHG emission reduction estimates after carbon capture and sequestration showed potential to reach carbon neutral for biogas production and utilization processes, with a surplus of negative GHG emissions in some cases. These negative GHG emissions can be applied to offset GHG emissions from other waste management processes for which no GHG emission reduction processes are either available, or require a long-term implementation period.[46] Curbing emissions from the waste management sector has been previously proposed as a significant contribution towards the global net-zero emissions goal.[47,48] However, while these studies are focused in the management of municipal solid waste (MSW), we explored the potential application of the surplus negative GHG emissions from some of the assessed scenarios towards the abatement of GHG emissions from the waste management sector in Canada.

Fig. 7 shows the 2021 provincial GHG emissions from a range of waste management activities provided by Environment Canada,[37] as well as the estimated GHG emissions after offsetting these emissions with the surplus negative GHG emissions achieved in Scenarios 1 to 3. A surplus of negative GHG emissions was only obtained in some Scenario 1 and Scenario 3 cases.

GHG emissions from provincial waste incineration could be completely offset in Manitoba, and Newfoundland and Labrador. Alternatively, the GHG emissions generated by the forest industries of Prince Edward Island and Manitoba could be compensated by their provincial WRRF carbon capture, or they could mitigate the GHG emissions from the biological treatment of solid waste in Newfoundland and Labrador. Although full GHG emission offset was not possible for any waste management activity in the other provinces, it is notable to remark that approximately half of the GHG emissions generated by biological treatment of solid waste or waste incineration could be offset in Ontario. RNG generation in Quebec represents a significant share of biogas utilization. As such, Quebec carbon capture estimates are negatively impacted by the lower carbon dioxide capture efficiency from RHG generation compared with carbon dioxide capture from combustion offgases. The estimated Quebec GHG emissions offset by the WRRF negative emissions is lower than for the other provinces, with less than 50% reduction for all waste management activities. The high carbon intensities associated with electric power production prevent the possibility of negative GHG emissions in Nova Scotia, Alberta, and Saskatchewan, and therefore the potential offset of any waste management activity in these provinces.

4 Conclusions

WRRF biogas production and utilization is an advantageous and common alternative for treating and recovering valuable resources from municipal sewage sludge. Although this approach aligns with the emerging paradigms of circular economies and sustainable systems, the estimates in this work demonstrate that biogas production and utilization is not carbon neutral due to the net GHG emissions from methane-rich biogas leaks. In this work, we explored the potential of carbon capture technologies applied to WRRF biogas generation, and used processes to estimate the generation of negative GHG emissions, while leveraging the biogenic origin of biogas

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generated from the organic matter contained in sewage sludge. Three scenarios were assessed for their GHG emission reduction potential, with uncertainty sources addressed with a Monte Carlo method model. Scenario 1 estimated the effects of technologically mature CCS processes capturing biogas combustion off-gases, or carbon dioxide separated after RNG production. Scenario 1 addressed the capture of both the carbon dioxide component of biogas and carbon dioxide produced from its methane combustion. Conversely, the in-digester carbon capture though mineralization assessed in Scenario 2 captured only the carbon dioxide content in biogas, but it also provided the benefits of a higher energy density biogas, and the reduction of biogas volume, and downstream biogas flowrates that would require smaller sized equipment and piping systems. Although in-digester carbon mineralization would significantly reduce GHG emissions from biogas production and utilization, its capture efficiency is too low to completely offset provincial GHG emissions from biogas production and use at WRRFs. The integration of both Scenarios 1 and 2 was explored under Scenario 3 to evaluate the synergetic effect of reducing the carbon dioxide content in biogas at the production point, which may result in smaller downstream biogas leak magnitudes, and the higher capture efficiency of implementing CCS systems after biogas combustion or upgrading. It was observed that the estimated carbon emissions from the carbon capture processes had significant impacts in those provinces with high carbon intensities for electric power generation, preventing the achievement of overall negative GHG emissions. Provincial WRRF biogas production and use may reach carbon neutrality for some cases under Scenarios 1 with the maximum reported carbon capture efficiency, and for all Scenario 3 cases.

In some of these carbon dioxide capture scenarios, a surplus of negative GHG emissions was generated. The potential of the surplus negative GHG emissions to offset GHG emissions from other waste management activities was evaluated. Although GHG emissions from landfill management and wastewater treatment—the largest provincial waste management processes—cannot be completely offset by WRRF CCS for any scenario in all Canadian provinces, GHG emissions from some smaller waste management processes, such as municipal solid waste or industrial wood waste were estimated to be completely offset in Manitoba, and Newfoundland and Labrador. Additionally, partial GHG emission offset by WRRF CCS for other waste management processes was estimated for other provinces and sectors, such as the biological treatment of solid waste or waste incineration in Ontario. This study demonstrates how fugitive GHG emissions from anaerobic digestion during municipal wastewater treatment reduce the sustainability of waste valorization processes, resulting in net GHG emissions. Consequently, strategies to abate or offset these GHG emissions must be developed and put into practice in order to achieve carbon neutrality, with the additional potential of a negative GHG emission surplus. In this regard, further work on the practical implementation of carbon dioxide capture and storage systems in processes using biogenic carbon is needed to assess their negative GHG emission production potential for sewage-treatment processes.

Author contributions

Edgar Martín-Hernández: conceptualization, data curation, formal analysis, investigation, methodology, visualization, writing – original draft, writing – review & editing. Kexuan Chen: investigation, formal analysis. Domenico Santoro: funding acquisition, writing – review & editing. Sidney Omelon: conceptualization, funding acquisition, supervision, writing – review & editing.

Conflicts of interest

There are no conflicts to declare.

Data availability

The datasets and scripts used in this study are available in the Zenodo repository, with the identifier https://doi.org/10.5281/zenodo.16342313.

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