1 2 3	Combined Heat and Power May Conflict with Decarbonization Goals - The Air Emissions of Natural Gas Combined Cycle Power versus Combined Heat and Power Systems for Commercial Buildings
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23 24 25 26 27 28 29 30 31	Abstract This study compares the environmental impacts of a centralized natural gas combined cycle (NGCC) versus a distributed natural gas-fired combined heat and power (CHP) energy system in the United States (U.S.). We develop an energy-balance model in which each energy system supplies the electric, heating, and cooling demands of 16 commercial building types in 16 climate zones of the U.S. We assume a best-case scenario where all the CHP's heat and power is allocated toward building demands to ensure robust results. We quantify the greenhouse gas (GHG) emissions, conventional air pollutants (CAPs), and natural gas (NG) consumption. In most cases, the decentralized CHP system increases GHG emissions, decreases CAP emissions, and decreases NG consumption relative to the centralized NGCC system. Only fuel cell CHPs were able to simultaneously reduce GHGs, CAPs, and NG consumption relative to the
33 34 35 36 37	NGCC-based system. The results suggest that, despite their energy efficiency benefits, standard distributed CHP-based systems typically do not have enough benefits versus an NGCC-based system to justify a reorganization of existing infrastructure systems. Since fuel cell CHPs can also use hydrogen as a fuel source, they are compatible with decarbonized energy systems and may aid in the transition towards a cleaner energy economy

Introduction

To mitigate the energy sector's environmental impacts, experts suggest increasing energy efficiency and decarbonizing our energy sources. Historically, debates regarding coal displacement from the United States (U.S.) energy mix suggested that highly efficient natural gas (NG) was a good alternative because it emits approximately half as much carbon dioxide (CO₂) as coal per unit of energy combusted. Between 2011 and 2019, 103 coal-fired power plants in the U.S. have been converted to or replaced by natural gas combined cycle power plants (NGCC) — which use steam and gas turbines to produce electricity. A. Since energy use to electrify and heat commercial & residential buildings constitutes over one-third of the U.S. greenhouse gas (GHG) emissions, distributed combined heat and power (CHP) systems may present an advantage over NGCC by improving the overall conversion efficiency.

CHPs couple a prime mover – the primary mechanical or electrochemical method of energy conversion – with a heat-recovery unit to recover and reallocate waste heat. Therefore, CHPs may provide an overall conversion efficiency that is higher than that of conventional thermal power plants (TPPs).⁶ Conventional TPPs are sized to supply the electricity needs of cities, consume water to reject heat,⁶ and are often situated outside of city limits – and power losses increase with transmission distance.⁷ We refer to this paradigm as 'centralized' power generation, in which a single power plant generates enough electricity to power a city. In comparison, CHPs are typically placed near the end-user to allocate recovered heat to satisfy the user's thermal demands (e.g., in large building complexes like hospitals and campuses).⁶ We refer to this paradigm as 'distributed' energy generation, where multiple generators can take the role of 'mini' power plants throughout a city to supply electricity and heat.

Previous studies suggest that the adoption of distributed CHPs may increase energy systems' resilience to blackouts or brownouts, ^{8,9} and reduce GHG and air pollutant emissions, ^{8,10–14} water-for-energy consumption, ^{10,11,14} and fuel consumption ^{13,15–17} associated with electricity generation when compared to the current U.S. electricity grid mix. Angrisani et al. studied the potential of internal combustion engines for a CHP system from a utilization rate, economic, and air pollution point of view. ¹⁷ Yan et al. developed a parametric cost-benefit model for a distributed CHP system and showed that the

lifecycle water consumption and sulfur dioxide emission are much lower for CHP compared to grid electricity. ^{14,18} Cappa et al. compared the technical and economic performances of a proton exchange membrane fuel cell to an internal combustion engine for CHP under a residential energy supply scenario. ⁹ A few studies have compared the life cycle impacts of a centralized NGCC to distributed CHPs but they do not consider how the energy outputs apply to the energy demands of buildings, which may limit how the energy outputs of CHPs are used. ^{15,16,19} For instance, CHPs can be coupled with absorption chillers to supply cooling loads to commercial buildings and industrial plants. ²⁰ Unlike electrically-driven air conditioners, absorption chillers are heat-driven systems that use a binary refrigerant-absorbent solution to absorb heat from hot water, steam, or combustion exhaust to chill water. ²⁰ Contrary to the cited studies above, Berrill and Hertwich found that the environmental benefits of CHP systems over separate heat and power systems in the U.S. are substantially dependent on the region, and may, in many cases, exacerbate GHG emissions. ¹⁹ Hence, more research is needed to identify when CHPs reduce the environmental burdens of building energy consumption.

In this article, we compare the air emissions of a hybrid energy system (HES) – which incorporates distributed CHPs and/or absorption chillers – to the lowest impact NG-based alternative that we refer to as the conventional energy system (CES). Our scope is limited to commercial building applications in 16 U.S. cities.²¹ We define the CES as an energy system that uses presently common technologies and system configurations. We developed an hourly energy balance simulation model for HES and CES to meet the build energy demand. Within the CES, electricity is supplied by an NGCC power plant and heat is supplied by a natural gas-fired furnace or boiler, and the cooling demand is met by an electric chiller. In practice, the CES would likely include a combination of renewable and non-renewable energy sources; thus, this is organized around a 'best case' for CHP to ensure robust results or a similar comparison. To compare the CES to the HES we calculate the operational GHG emissions (i.e., CO₂, methane (CH₄), and nitrous oxide (N₂O)), conventional air pollutant emissions (i.e., carbon monoxide (CO), nitrogen oxides (NO_X), particulate matter (PM), sulfur dioxide (SO₂), and volatile organic compounds (VOC)), and NG consumption.

Materials and Methods

91 System configuration

Figure 1 shows the system configuration for our CES and HES. In the CES, electricity, heating, and cooling are supplied by an NGCC plant, an NG-fired boiler, and an electrical air chiller (using electricity from the NGCC)), respectively. The HES is similar, but a fraction of the electricity is supplied by a CHP, the heat from the CHP substitutes the boiler heat to the extent such heat is available, and heat from the CHP is also utilized for an absorption chiller (ABC) that supplies the cooling. The adoption of a CHP or ABC does not necessarily substitute its counterpart. For instance, a building with a CHP may still draw electricity from the grid (which we assume is electrified by an NGCC) or heat from an onsite natural gasfired boiler in cases where the CHP does not provide enough energy to satisfy the building loads. Accordingly, we assume that buildings are connected to a CHP network, where a single CHP supplies heat and electricity to multiple buildings. Although the investment cost for redundant CES and HES technologies may be relatively high, the O&M cost can be reduced if the building reduces its use of the electric chiller and NG-fired boiler. Since existing commercial buildings may already have CES infrastructures, the investment cost of refurbishment to HES infrastructures may be economically impractical. Nonetheless, we limit our study scope to the environmental impacts of the CES and HES.

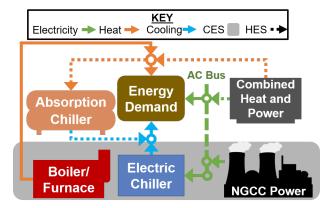


Figure 1. System configurations of the conventional energy supply (CES) – highlighted in the gray boundary – and the hybrid energy supply (HES) which can include all the technologies that are shown. The solid arrows represent the energy flows from conventional technologies and the dotted arrows represent energy flows from alternative technologies. A larger version of this figure is available in the supplementary information (see Figures S3 and S4).

Assumptions and Data Sources

Cities and Buildings. We used the U.S. Department of Energy (DOE) simulated demand energy profiles for 16 commercial reference buildings in each of the 16 representative cities of the U.S – which represent one of the 16 climate zones in the U.S.²² These energy demand profiles were generated through a collaborative effort of the DOE and three of its national laboratories in 2011 – see Deru et al (2011) for a detailed methodology.²¹ Generic attributes from each city are presented in the supplementary information (S.I.) (Table S1, and Figures S1 and S2). As of 2011, these commercial reference buildings represented the characteristics of over 60% of the commercial building stock in the U.S. and served as hypothetical models of their ideal operation.²¹ We assume that all building energy demands match the commercial reference building models,²¹ and adopt the NREL simulated demand reference period of 2004 – 2014.

Conventional Energy System. The CES supplies energy through grid-level electricity, an NG-fired boiler, and an electrical air chiller (AC). We assume that all grid electricity is supplied by an NGCC because it is the most efficient fossil fuel-fired centralized power plant that uses the same fuel as a CHP, thus, we compare CHP performance to the likely-best-case alternative NG-performance. Moreover, when all emissions are allocated to electricity production, NGCCs emit less CO₂ than most CHPs per unit of electricity generated.^{6,23}

The performance metrics for the NGCC were gathered from the 2020 Annual Technology Baseline²³ and the Argonne National Laboratory (Table S2).²⁴ The performance metrics of conventional heating and cooling equipment were obtained from the U.S. Energy Information Administration (EIA)²⁵ and Deru and Torcellini (see tables S3 and S4).²⁶ We assume that the buildings use a base-model commercial rooftop air conditioner with a coefficient of performance (COP) of 3.4 and an NG-fired boiler with a 99% efficiency.

Hybrid Energy System. The HES supplements the CES by adding a CHP and/or an absorption chiller (ABC) (Figure 1). The EPA catalogs the performance of five CHP classes: (1) reciprocating engine, (2) gas turbines, (3) steam turbines, (4) microturbines, and (5) fuel cells.⁶ The rates of fuel consumption, electricity and heat generation, and emissions outputs depend on the thermodynamic cycle

of each CHP (see Figure S3 and Table S6). Steam turbines were excluded in this analysis because they are typically limited to industrial applications rather than commercial applications – our study's focus. For model simplification, we assume that each CHP operates at capacity and has a constant emission and NG consumption rate, efficiency, and heat-to-power ratio (HPR) (Table S6). This assumption is fairly optimistic since it ignores the fact that there may not be any demand for the generated heat which would reduce our overall system efficiency. Rather, our simulations represent a 'best-case scenario' in which all of the energy is allocated to the commercial buildings' energy demands and indicate the upper limit for the environmental benefits of our HES.

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We assume that ABCs couple with either the CHP or the onsite NG boiler to assess the benefits and drawbacks of each technology. In cases where a CHP or other primary heat source cannot supply the ABC with the amount of heat demanded, an auxiliary boiler can be used to supply hot water to the ABC. ^{27,28} Previous studies assume that CHPs and ABCs are coupled (i.e., combined cooling, heating, and power – or CCHP) to exploit the CHP's heat output during warmer months. However, this approach obscures the added benefits of each technology. By decoupling the CHP and the ABC, we implicitly question whether NG electricity or heat is better to cool buildings. ABCs' viability is typically dismissed compared to electric chillers because they have a lower coefficient of performance (COP) and have higher capital costs.²⁹ These concerns might not reflect environmental and economic performances because 1) ACs do not consider upstream energy losses in the COP calculation; and, 2) ABCs may lead to a quicker or comparable payback period through lower operating costs.²⁹ It is worth noting that electrifying all building services – including heating and cooling – has the advantage of enabling cleaner future conditions as the grid decarbonizes, which is not true for a natural gas CHP heat-dependent infrastructure. We use a single-stage ABC with a COP of 0.7. We exclude two-stage ABCs (COP \geq 1.35) because we do not model the type of heat output from the CHPs (i.e., hot water, steam, or direct exhaust) and this is beyond our present scope. A two-stage ABC would likely reduce the amount of heat required to cool a building relative to the single-stage ABC and this assumption limits the robustness of our general results.

- In cases where the CHP cannot supply the building's heat demand, an onsite NG boiler supplies the remaining heat demand.
- 165 Modeling

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- For a full description of the energy balance model and calculations, see Section 2 of the S.I.
- Supply and Demand. We aggregate the hourly building energy demands into either electricity (E), heating
- 168 (Q), or cooling (C). Technologies i, j, and k supply electricity, heating, and cooling, respectively, at each
- timestep as shown in equations (1) through (3).

$$E_D = \sum_{i=1}^{\infty} E_i \tag{1}$$

$$Q_D \le \sum_{j=1} Q_j \tag{2}$$

$$C_D = \sum_{k=1}^{\infty} C_k \tag{3}$$

- By inequality (2) our system may generate more heat than is required to meet the heat demand of a building. For instance, a CHP with a high heat-to-power ratio may produce more heat than is required for the building when supplying the electricity demands at a given time. In these cases, we assume that surplus heat is reallocated to other buildings. Depending on the distributed heat network, flow media, and insulation properties, distributed heating and cooling systems may lose between 1% and 34% of their energy in distribution.^{30–32} In keeping with our interest in realistic but best-case CHP conditions, we use the Swedish average distribution loss of 10% distribution loss for all heating and cooling supplied by the CHP and the ABC.^{30,31} A longer discussion on energy losses for distributed energy systems can be found in Section 3.6 of the SI. We do not consider electric nor thermal energy storage systems within our study scope.
- Impacts and Efficiency. We calculate the annual emissions for two air pollutant classes: GHGs i.e.,
 CO₂, CH₄, and N₂O and conventional air pollutants (CAP) i.e., CO, NO_X, PM, SO₂, and VOCs. We
 use this classification because each pollutant has different scales of impacts. GHGs have long-term and

global impacts while CAP emissions contribute to acute and localized public health impacts (i.e., NO_X and VOCs are precursors to tropospheric ozone and smog formation). We exclude non-operational GHG and CAP emissions because previous articles show that the equipment's manufacture contribution towards the overall life cycle impacts was negligible compared to the other life cycle stages – i.e., operational emissions, fugitive emissions, and fuel acquisition & delivery (FA&D) – of NG-fired generation systems. 15,16

We calculate the total emissions as the sum of the operational and leakage emissions of pollutant *x* minus the avoided emissions (discussed below) using equation (4). We use the average CH₄ leakage rate estimated by Alvarez et al. (2.5%),³³ which represents the cumulative mass fraction of CH₄ leaked from production through distribution of the US oil and NG system normalized by the gross mass of CH₄ withdrawals.^{33,34} We adjust the leakage rates estimated by Alvarez et al.³³ to represent the fraction CH₄ leaked normalized by the gross NG withdrawals by assuming that NG is 90% methane by mass (Table S7 and S8).³⁵ The NG system can be subdivided into four stages: *production*, *processing*, *transmission*, and *distribution*. For leakage values associated with the NGCC, we exclude the *distribution* stage since NG-fired power plants are supplied directly by the transmission pipelines from the NG processing and treatment plants.³⁴ We assume that the boilers' and CHPs' NG supply is delivered through the NG *distribution* system, which accounts for approximately 0.11% of methane leaks normalized by the gross natural gas withdrawals (Table S8).³⁴

We assume that any surplus heat produced by the CHP is reallocated to meet heat demands in other buildings that would otherwise have been provided by an NG-fired boiler. Accordingly, we assign CHP emissions or fuel consumption credits (CHP credits) since reallocating CHP heat offsets heat generation from an NG-fired boiler. The CHP credits are the avoided impacts equivalent in mass to the emissions or fuel that would otherwise be emitted or consumed by the NG-fired boiler to produce the equivalent amount of heat. Regarding CH₄, the CHP credits include offsets for avoided leakage (see equation S26 in the S.I.). For instance, a commercial gas boiler produces 0.182 kg CO₂ per kWh_{th} of heat (Table S3); thus, 10 kWh_{th} of surplus heat would displace 1.82 kg of CO₂. Because the FA&D phase

contributes significantly towards the life cycle impacts,¹⁵ we calculate the NG (fuel) consumption of our system (*F*) using Eq. 5. In calculating fuel consumption (Eq. 5), we only consider the NG consumed onsite and exclude the leakage of NG because the leakage reflects inefficiencies of the NG system more so than those of the energy generation methods.

$$e_x = e_{x,energy\ generation} + e_{x,leakage} - e_{x,CHP\ credit}$$
(4)

$$F = F_{NGPP} + F_{CHP} + F_{Furnace} \tag{5}$$

We adapt the Total Fuel Cycle Efficiency (TFCE) metric from Best et al., ¹³ (Eq. 6) to quantify the overall system efficiency. The TFCE equals the useful energy output (i.e., electricity and heat) divided by the net energy input (i.e., NG). Similarly, we can quantify the amount of useful energy as the difference between the energy input and the total wasted heat by the system (Eq 7).

$$TFCE = \frac{\sum Useful \ E \ and \ Q}{F}$$
 (6)

$$TFCE = 1 - \frac{Q_{waste}}{F} \tag{7}$$

Results and Discussion

We plot the emissions and fuel consumption values against the annual energy demand intensity (AEDI), defined as the sum of the annual electricity ($E_{D,Annual}$) and heating demands ($Q_{D,Annual}$) divided by the reference building's floor area. The AEDI refers to the required energy supply of a building to satisfy its heating, cooling, and electrical loads. The AEDI for each building in each climate zone is shown below (Figures 2a and 2b). We choose the AEDI because it includes each building's electricity, heating, and cooling demands for its respective climate zone (see SI section 3.1 and Figures S2 to S4).

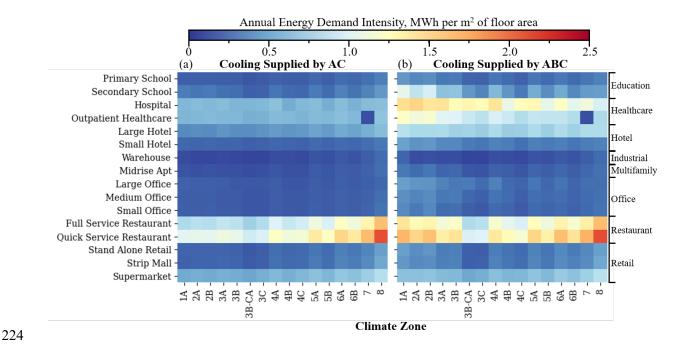


Figure 2. Annual energy demand intensities (MWh m-²) for 16 commercial reference buildings in the 16 climate zones of the U.S. (a) shows the annual energy demands when all the cooling is supplied by an electrically powered cooling system (i.e., an AC system). (b) shows the annual energy demands when all the cooling is supplied by a heat-powered cooling system (i.e., an ABC). We exclude region 7's outpatient healthcare building simulation results because its energy demands appear to deviate significantly from the other regions (See SI section 3.1).

Greenhouse Gas Emissions

Figure 3 shows the GHG emissions intensity (t CO_2 eq per m^2 of floor area) for all the buildings simulated when the CHP supplies all the electricity demand. The black line serves as our reference datum, which represents the emission values of the CES (i.e., NGCC electricity, NG-fired boiler, and an AC system for cooling). To aggregate the GHG emissions, we used the 100-year global warming potential (GWP₁₀₀) for CH₄ (28) and N₂O (265).³⁶ The GWP₁₀₀ values we used from the Intergovernmental Panel on Climate Change Synthesis Report (AR5) do not include climate change feedbacks (e.g., the increase in global temperatures caused by CH₄ emissions may subsequently increase the amount of water vapor in the atmosphere). We also provide the unaggregated emissions of CH₄, CO₂, and N₂O and the relative change for all GHG emissions in Figures S8 – S14.

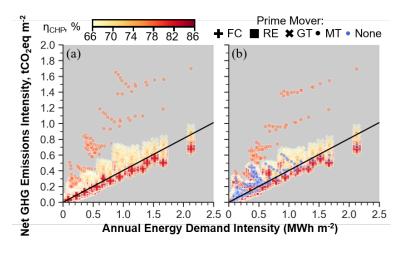


Figure 3. The annual GHG emissions intensity (tonnes $CO_2eq\ m^{-2}$) for all simulated buildings when the CHP supplies all the electricity demand and the cooling is supplied by (a) an AC system or (b) an ABC system. The black line represents the best-fit line of the reference datum emissions for the CES ($R^2 = 0.96$). The color gradient represents the CHP efficiency (η_{CHP}) of the prime mover, and the marker type indicates the type of prime mover used. The blue markers indicate that an ABC was adopted without being coupled to a CHP.

Relative to the CES, the HES, on average, reduces the total N₂O emissions but increases the total CO₂ and CH₄ emissions. N₂O is usually emitted by combusting nitrogen-bonded fuels and not for the combustion of NG and biogas.³⁷ Accordingly, we assumed that N₂O emissions from CHP systems were negligible, resulting in a significant drop in the total N₂O. Only fuel cells and reciprocating engines reduce CO₂ emissions when coupled with an AC system – otherwise, CO₂ emissions increase. The fuel cell and reciprocating engine classes had the lowest CO₂ emissions factors and were on the higher range of the efficiencies, reaching greater than 80% conversion of the NG into useful energy. CH₄ emissions increase in most cases, which we discuss in the 'Leakage' section below. The increase in GHG emissions relative to the CES is driven by the CHP's CO₂ emissions and by CH₄ emissions from increased onsite heating and leakage – especially when an ABC is adopted (Figures S10 and S14). These results are consistent with previous literature comparing NGCC and distributed CHP systems.^{15,16} When the disparity between the GHG emissions profiles of the AC and ABC scenarios is large (Figure S17), the CHP is not generating enough heat and cannot supply the increased requirement for heat to satisfy the heating and cooling loads of the building (Figure S8).

displaces the NGCC for electricity supply. Fuel cells – which constitute 22% of the cases (2550 of 11,475) – reduced GHG emissions in 88% of cases (1119 of 1275) with AC cooling and 36% of cases (457 of 1,275) with ABC cooling. Other CHP classes may also reduce GHG emissions relative to the CES at lower levels of service – i.e., CHP and the NGCC both supply a fraction of the electricity; however, we do not explore the relationships between all the variables or present this here due to space limitations. Relative to the CES, the AC-CHP system increases GHG emissions for all combustion- CHPs – reciprocating engines ($2\% \pm 6\%$), gas turbines ($28\% \pm 13\%$), microturbines ($60\% \pm 72\%$) – whereas fuel cells, on average, reduce the relative GHG emissions by $7\% \pm 5\%$. In cases where we show negative absolute emissions (e.g., negative tCO₂ m⁻²) we mean the system may offset the GHG emissions from an NG-fired boiler, not that CHPs are carbon negative. Similarly, most CHPs increase GHGs relative to the CES when the ABC system supplies the cooling – fuel cells (11 $\% \pm 7\%$), reciprocating engines (14 $\% \pm$ 18%), gas turbines (34% \pm 16%), and microturbines (58% \pm 58%). Leakage Methane emissions from leakage contribute, on average, more than 30% of the total GHG emissions. The CES leakage emissions (38 \pm 31 kg CO₂eq per m² of floor area) are approximately 36% \pm 2.4% of total GHG emissions. Table 1 shows the average emissions from leakage and the relative change of leakage emissions relative to the CES when the CHP supplies all the electricity demand. The ABC system increases onsite heat demand, onsite NG demand, and subsequently, leakage from the NG system. The contributions of fugitive emissions to GHG emissions is consistent with previous studies. 15,16 If we assume no leakage, the results are fairly similar in that only fuel cells and reciprocating engines reduce operational GHG emissions relative to the no-leakage CES with both cooling systems. Specifically, GHG emissions decrease relative to the no-leakage reference datum in 41% of cases (2,279

Relative to the CES, all CHPs except fuel cells increase GHG emissions when the CHP fully

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of 5,610) with AC cooling and 20% of cases (1,134 of 5,610) with ABC cooling.

Table 1. The average GHG emissions (kg CO₂eq m⁻²) from leakage and relative change of methane emissions from leakage (%) relative to the CES leakage emissions— in parentheses— when the CHP system supplies all the electricity demand.

Prime	Cooling Scenario							
Mover*	AC	C (%)	ABC (%)					
None	38 ± 31	0	46 ± 36	(24 ± 21)				
FC	41 ± 34	(10 ± 18)	45 ± 35	(22 ± 21)				
RE	46 ± 38	(24 ± 21)	48 ± 38	(31 ± 20)				
GT	56 ± 46	(52 ± 26)	56 ± 45	(51 ± 21)				
MT	57 ± 46	(56 ± 23)	56 ± 45	(52 ± 19)				

*Note: FC = Fuel Cell; RE = Reciprocating Engine; GT = Gas Turbine; MT = Microturbine

Global Warming Potential

Over the past decades, the Intergovernmental Panel on Climate Change's GWP estimates for methane increased and may continue to do so in the future. The GWPs we used neglect climate-carbon feedbacks that would otherwise increase the 100-year GWP (GWP $_{100}$) of methane and nitrous oxide by 21% and 12%, respectively. Our analysis shows that a 10% change to methane's GWP $_{100}$ corresponds to an average 3.7% change in overall GHG emissions (Table S9). Relative to the CES, sensitivity to the GWP $_{100}$ is lower for all combustion-based CHPs and higher for fuel cells. Initially, this might suggest that at higher GWPs, CHPs are more beneficial since they offset more heat generation from NG-fired boilers. However, under the 20-year GWPs (CH $_4$ – 84 and N $_2$ O – 264) $_3$ 6 – a 200% increase in methane's GWP $_{100}$ values - only fuel cells reduce GHG emissions relative to GWP $_{20}$ CES. Under the best-case scenario (CHP with AC cooling), GHG emissions drop in 17% of the cases (976 of 5,610), which is approximately half of our GWP $_{100}$ results (1,785 of 5,610). Accordingly, the GWP values are heavily influential in the decision criteria as methane leakage has a higher weight. These results indicate that combustion-based CHPs are generally ineffective as an NGCC replacement to reducing GHG emissions for commercial buildings, and agree with Berril and Hertwich, who indicate that CHPs do not necessarily reduce GHG emissions relative to the existing energy infrastructure for commercial buildings.

Conventional Air Pollutants

We quantified the change in CO, NO_X, PM, SO₂, and VOC emissions. In cases where we show negative absolute CAP emissions (e.g., negative g m⁻²), we mean the system may offset the CAP emissions from an NG-fired boiler, not that CHPs are absorbing CAP emissions. The dominant drivers for CO and NO_X emissions are heat demand and supply, and electricity demand and supply are the dominant drivers for PM and SO₂ emissions. Figure 4 below shows the annual NO_X emissions intensity for all buildings simulated. The CO emission trends (Figure S20) are very similar to those for NO_X (Figure 4). The NG-fired boiler's CO emissions factor is approximately four times larger than the NGCC, while the NO_X emission factor is nearly three times larger (Tables S2 and S3). Compared to the NG-fired boiler, most CHPs have smaller NO_X emissions factors (Table S6). When paired with an AC system for cooling, CHPs offset heat generation from an NG boiler and subsequently reduce the overall emissions of CO and NO_X in over 99% of our cases. The coupled CHP and ABC may increase CO and NO_X emissions where the CHP cannot meet the elevated heat demands (Figure S7). In these cases, the CCHP combo reduces CO and NO_X in over 70% of the cases. CHPs with high HPRs and fuel cells reduce CO and NO_X the most because more NG-boiler heat is offset, and they have negligible CO and NO_X emissions, respectively.

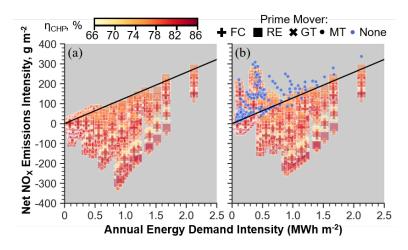


Figure 4. The annual NO_X emissions intensity (g m⁻²) for all simulated buildings when the CHP supplies all the electricity demand, and the cooling is supplied by (a) an AC system or (b) an ABC system. The black line represents the best-fit line of the CES' NO_X emissions for the CES ($R^2 = 0.98$). The color gradient represents the CHP efficiency (η_{CHP}) of the prime mover, and the marker type indicates the type of prime mover used. The blue markers indicate that an ABC was adopted without a coupled CHP.

SO₂ and PM emissions are dependent on the fuel type used in combustion systems. SO₂ emissions depend on the sulfur content of the fuel are usually associated with liquid, solid, and landfill gas fuels.⁶ In the case of CHP systems, sulfur can degrade the CHPs' performance and decrease their lifespan, especially in gas turbines and fuel cells.⁶ To mitigate these losses, sulfur is typically removed before NG is fed to the CHP using sulfur adsorbent bed catalysts.⁶ PM emissions are a carryover from non-combustible constituents in the fuel or the incomplete combustion of larger molecular weight hydrocarbons.³⁵ Accordingly, we assume the only CHP systems undergo a desulfurization process before combustion and that PM and SO₂ are primarily emitted by the NGCC and the NG-fired boiler.⁶ Because, the NG-fired boiler has a lower emissions factor for both PM and SO₂ than the NGCC (Tables S2 and S3), adopting an ABC for cooling also reduces their emissions relative to the baseline. The adoption of a CHP displaces the emissions from both the NGCC and the NG-fired boiler and will always reduce PM and SO₂ emissions (Figures S32 through S24). Since surplus heat from the CHP will offset heat from another NG-fired boiler, CHPs with higher HPRs produce the largest reductions of PM and SO₂.

The assumption we made for CHP's SO₂ and PM emissions is based on our literature review and the lack of data inventory, which presents a severe limitation. The disparity of inventory data regarding the SO₂ and PM emissions of NGCCs and CHPs may also be a result of the monitoring programs within the EPA's Clean Air Markets Division. Our source for the NGCC's SO₂ emissions factor can be traced from NREL to the EIA, to the EPA. Further inspection of the EPA's Emissions & Generation Resource Integrated Database, or eGRID, reveals that out of the 6802 NG units reporting emissions data, over 70% estimate their SO₂ emissions, while the others do not report a source.⁷ Accordingly, it is quite possible that the difference in SO₂ emissions between the NGCC and CHP is negligible. Nonetheless, NGCC power plants are still required to report SO₂ emissions through the Acid Rain Program. For context, an NGCC produces ~100 times less SO₂ than coal-fired power plants.³⁸

In most cases, VOC emissions increase when a CHP supplies electricity, and when ABC supplies the cooling (Figure 5). Compared to an NGCC, the VOC emission factor of all CHPs is an order of magnitude greater − except for fuel cells and a couple of microturbines with negligible VOC emissions (≤

0.01 kg MWh⁻¹, see Table S6). Accordingly, only these fuel cells and microturbines reduce VOC emissions relative to the baseline when the AC system supplies the cooling (Figure 5a). When the ABC system supplies the cooling, only fuel cells reduce emissions below the baseline in cases where the CHP generates surplus heat. Accordingly, we find that the dominant driver of VOC emissions is the method of energy generation (i.e., combustion vs. electrochemical) and emissions control. Fuel cells combust the off-gasses with plenty of excess air (lean-combustion) at temperatures high enough to oxidize VOCs and CO, while simultaneously preventing NO_X formation (less than 1,800°F). The remaining CHPs use a catalytic process to oxidize VOCs, CO, and NO_X.

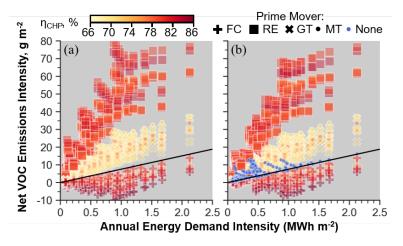


Figure 5. The annual VOC emissions intensity (g m⁻²) for all simulated buildings when the CHP supplies all the electricity demand, and the cooling is supplied by (a) an AC system or (b) an ABC system. The black line represents the best-fit line of the baseline emissions for the CES ($R^2 = 0.98$). The color gradient represents the CHP efficiency (η_{CHP}) of the prime mover, and the marker type indicates the type of prime mover used. The blue markers indicate that an ABC was adopted without a coupled CHP system. The relative change in VOC emissions can be found in the SI (Figure S27).

Natural Gas Consumption and Total Fuel Cycle Efficiency

Figure 6 shows the NG consumption for each system. These consumption values only consider the operational use of NG and do not include the NG lost between the production and distribution stages of the NG system. Relative to the CES, NG consumption decreases in 12% of cases (673 of 5610) with AC cooling (Figure 6a) and 5% of cases (306 of 5610) with ABC cooling (Figure 6b). Most of the reductions in NG consumption can be attributed to fuel cells (80% of cases) and reciprocating engines (20% of

cases). Microturbines did not reduce NG consumption and gas turbines reduced NG consumption in only 1 scenario.

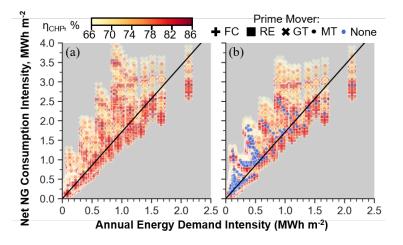


Figure 6. The annual NG consumption intensity (kWh m⁻²) for all simulated buildings when the CHP supplies all the electricity demand and the cooling is supplied by (a) an AC system or (b) an ABC system. The black line represents the best-fit line of the baseline emissions for the CES ($R^2 = 0.92$). The color gradient represents the CHP efficiency (η_{CHP}) of the prime mover, and the marker type indicates the type of prime mover used. The blue markers indicate that an ABC was adopted without a coupled CHP system. The relative change in NG consumption can be found in the SI (Figure S28).

The combustion compressors along the pipeline produce the most upstream emissions in the NG system's lifecycle due to inefficiencies and incomplete combustion. Although we do not calculate the FA&D-associated emissions, Mann et al. Peport that the FA&D stage contributes to the CHPs' lifecycle GHG (< 20%), CO (> 60%), NO_X (> 80%), and VOC ($\sim 20\%$) emissions assuming a CH₄ leakage rate of $1.4\% \pm 0.5\%$ of the gross NG produced. Since this leakage rate is understood to be underestimated, we expect the contributions of FA&D towards the overall emissions to be higher. More research is needed to quantify how reducing NG consumption in the HES impacts its overall lifecycle emissions.

To better observe trends and changes to the total fuel cycle efficiency (TFCE), we separated them by building types (Figures S27 – S37). We used logarithmic regression to generate the best fit line for the CES (Table S10). In most cases, the addition of a CHP or an ABC increases the TFCE by the virtue that they decrease the amount of wasted energy (i.e., waste heat from the NGCC). Accordingly, the TFCE may increase regardless of increased or decreased NG consumption. Rather, higher TFCE values are found alongside higher efficiency CHPs (Figures S29-S39). On average, a CHP and an ABC increase the

TFCE by the virtue that more of the combustion energy is allocated towards the demands. On average, the TFCE increased by $24\% \pm 16\%$ relative to the baseline in 97% (5,441 of 5610) and 98% (5524 of 5610) of the cases for AC and ABC cooling, respectively. Fuel cell CHPs produced the highest TFCE values – 87% and 92% for AC and ABC cooling, respectively. These results conform with a review by Elmer et al., 39 who indicate that the overall system efficiency for fuel cell tri-generation systems (i.e., fuel cell CCHPs) ranges from 72% with molten carbonate fuel cells to 87% with solid oxide fuel cells.

Nonetheless, we identify a major drawback of the TFCE metric. For example, a scenario where a building changes from an AC to an ABC without adopting a CHP may increase the amount of energy consumed on-site, as previously discussed. However, this transition reduces the amount of wasted energy from the NGCC since the heat for the ABC is supplied by the NG-fired boiler. In this scenario, NG consumption and the TFCE increase. If the goal of the decision-maker is to reduce NG consumption, the TFCE by itself may misinform stakeholders.

Comparison

 Compared to a CES that uses an NGCC for power generation, only fuel cell CHPs with a CHP efficiency greater than 80% improved on all the metrics considered in this paper. For context, the CHP efficiency for fuel cells ranges from ~75% to 90%. ^{6,39} Since fuel cells can also operate hydrogen and biogas, they may reinforce a renewable energy transition especially if hydrogen is used as the energy storage medium for intermittent solar and wind power. ⁴⁰ To improve the performance of distributed CHP systems, one may instead try to match the community profile to the CHP energy outputs. ¹³ A stronger focus should be placed on evaluating CHPs for district heating and cooling applications. We summarize our results in Table 2 below.

Table 2. Average relative change for the adoption of CHPs and absorption chillers, relative to a conventional energy system with an NGCC. A detailed summary for each CHP is available in Table S11 of the SI.

СНР		GHGs Criteria Air Pollutants						NG	TECE	
Class	CO ₂	CH ₄	N_2O	СО	NOx	PM	SO_2	VOC	Cons.	TFCE
FC	-12*	10	-102	-133	-173	-114	-102	-111	7	33
GT	21	51	-107	-141	-209	-133	-106	165	47	13

MT	64	55	-109	-172	-241	-143	-108	110	51	18	
RE	-5*	23	-105	-72	-156	-126	-104	666	21	30	
ABC	25	24	-7	76	153	29	-8	72	23	21	

FC: Fuel Cell, GT: Gas Turbine, MT: Microturbine, RE: Reciprocating Engine, ABC: Absorption Chiller

Limitations and Uncertainties

The study has a few limitations and uncertainties. First, we only investigate NG-based systems. By comparing a CHP-based system to an NGCC-based system, we exclude advances in decarbonization, which may make CHPs irrelevant. By opting for a distributed CHP infrastructure, cities would commit to a new energy infrastructure that is less compatible with an energy supply that is becoming 'cleaner' over time (i.e., the electricity grid). If renewable energy systems use hydrogen-energy storage, fuel cell CHPs may be an exception because they use hydrogen to generate power. The fact that the distributed CHP-based system we studied does not necessarily perform well relative even to the NGCC-based system suggests there are not enough benefits versus an even cleaner energy system to justify a massive reorganization of infrastructure systems, ongoing investments, or decarbonization time frames.

Second, methane leakage is highly uncertain and varies spatially. ^{33,34,41} Since methane emissions from leakage contribute more than 30% towards GHG emissions in our study, the regional variability and uncertainty may certainly bolster or weaken the case for distributed CHPs. A commitment to a distributed CHP infrastructure would require increased oversight and mitigation strategies for methane leakage. We are unsure how these strategies would impact the overall lifecycle impacts or costs. The uncertainty of methane's GWP compounds leakage emissions' impact on GHGs. ³⁴ As we demonstrated, the choice of the 20- or 100-year GWP estimates significant impacts on the total GHG emissions and the perceived performance of CHPs and the NGCC.

We use the model developed in our study to assess the environmental impacts of centralized versus decentralized NG systems in the U.S. However, the implications of this study are more valuable in

 $[\]uparrow$ or \downarrow : Average trend when CHP supplies electricity and an AC supplies the cooling load.

^{*:} Indicates whether the average trend may reverse when an ABC supplies the cooling load.

countries with fossil fuel-based district heating systems, whose electricity mix remains highly dependent on fossil fuel, and may not rapidly transition to renewable energy (e.g., Russia, China, India, and Poland). 42-44 For instance, fossil fuels are 98% of Russia's energy supply – of which 75% is NG – and China is rapidly displacing coal with NG. 42,43 This study may also be relevant for developing nations in Africa that still heavily dependent on fossil fuels (e.g., coal, gas, diesel, and paraffin) and may not have enough capital to invest in city-level infrastructures. 45 This model can inform policymakers, researchers, and energy system planners alike on the benefits and drawbacks of CHP systems given our outlined assumptions and limitations. Since one of the major benefits of CHP is offsetting heat generation from other systems in commercial buildings, and previous work has also shown that CHP with thermal storage has potential for GHG emissions, 19 more research is needed for improved heat dispatch strategies or thermal storage options. Finally, this model could be improved by analyzing the interactions between a distributed CHP, renewables, and storage systems since previous work has suggested CHP-PV systems can reduce the life cycle impacts of the existing grid mix. 18

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487 **Associated Content**

- The supporting information (S.I.) is available free of charge on the ACS Publications website at DOI:
- 489 **(insert hyperlink here).** The S.I. further discusses the data inputs for our simulation, the derivation of
- our model, and more detailed results. A repository with the code used for the simulation can be found at
- 491 https://github.com/BBISSlab/DistributedEnergyGen.

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