1 Title

- 2 Benchmarking Greenhouse Gas Emissions from U.S. Wastewater Treatment for Targeted
- 3 Reduction

5 Authors

4

11

- 6 Sahar H. El Abbadi^{1,#,*}, Jianan Feng^{2,#}, Abigayle R. Hodson¹, Maryam Amouamouha³, Margaret
- 7 M. Busse⁴, Christina Polcuch¹, Pengxiao Zhou⁵, Jordan Macknick⁶, Jeremy S. Guest^{2,7}, Jennifer
- 8 R. Stokes-Draut¹, Jennifer B. Dunn^{5,8}
- 9 #Indicates equal contribution
- 10 *Corresponding author: <u>elabbadi@lbl.gov</u>

12 Affiliations

- 13 Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley CA 94720, United States
- ²The Grainger College of Engineering, Department of Civil and Environmental Engineering,
- 15 University of Illinois Urbana-Champaign, Urbana, IL 61801, United States
- 16 ³Loyola University Chicago, 305 Cuneo Hall, 1032 W. Sheridan Rd., Chicago IL 60660, United
- 17 States
- ⁴Department of Mechanical Engineering, Pennsylvania State University, University Park, PA
- 19 16802, United States
- ⁵Department of Chemical and Biological Engineering, Northwestern University, Evanston IL
- 21 60208, United States
- ⁶National Renewable Energy Laboratory, 15013 Denver West Parkway, Golden, CO 80401,
- 23 United States
- ⁷Institute for Sustainability, Energy, and Environment, University of Illinois Urbana-Champaign,
- 25 Urbana, IL 61801, United States
- ⁸Center for Engineering Sustainability and Resilience, Northwestern University, Evanston IL
- 27 60208, United States

29 Abstract

- 30 To assess the national climate impact of wastewater treatment and inform decarbonization, we
- 31 assembled a comprehensive greenhouse gas inventory of 15,863 facilities in the contiguous United

States. Considering location and treatment configurations, we modeled on-site CH₄, N₂O, and CO₂ production and emissions associated with energy, chemical inputs, and solids disposal. Using a Monte Carlo simulation, we estimate median national emissions are 47 million tonnes CO₂-eq·year⁻¹, with onsite process CH₄ and N₂O emissions 41% above current governmental estimates. Treatment configurations with anaerobic digesters are responsible for 16 million tonnes CO₂-eq·year⁻¹ of fugitive methane, outweighing benefits achieved through on-site electricity generation. Systems designed for nutrient removal have the highest greenhouse gas emissions intensity, attributable to energy requirements and N₂O production, demonstrating current tradeoffs between meeting water quality and climate objectives. We analyzed key sensitivities and included a geospatial analysis highlighting scale and distribution of opportunities to reduce life cycle greenhouse gas emissions.

Main

Wastewater treatment is essential for protecting public health and the environment. However, treatment processes generate greenhouse gases (GHGs) while relying on energy and chemicals whose production also contributes to total emissions.¹ Globally, the Intergovernmental Panel on Climate Change (IPCC) estimated wastewater treatment emissions to be 0.38 Gt CO₂-eq in 2019, comparable to key industries targeted for decarbonization, including the chemical (0.37 Gt CO₂-eq), cement (0.82 Gt CO₂-eq), and iron and steel (1.35 Gt CO₂-eq) industries.² Yet, recent studies found IPCC and U.S. Environmental Protection Agency (U.S. EPA) may underestimate methane (CH₄) emissions from on-site processes alone by two-fold,^{3,4} and nitrous oxide (N₂O) emission factors are often oversimplified and/or inaccurate.⁵ This study aims to improve understanding of wastewater GHG emissions, which are critical for meeting global climate targets as this sector grows with increasing population and expansion of essential sanitary services.

On-site emissions at wastewater treatment plants (WWTPs, also called water resource recovery facilities) are dependent on wastewater characteristics (e.g. organics, nitrogen), treatment processes, level of treatment, and plant size.^{3,5} N₂O emissions span multiple orders of magnitude, but appear correlated with treatment objective.^{5,6} Biogas used for on-site power generation can reduce imported energy, but leaking equipment and treated solids handling emits CH₄.³

Additionally, upstream electricity emissions depend on power requirements of treatment processes and regional grid carbon intensity.

Given this complexity, emissions reduction strategies must consider key plant and geospatial characteristics, energy, and material inputs, factors not currently accounted for in national emissions inventories.⁷ Internationally, several studies report national wastewater treatment inventories (Supplementary Table S1), including on-site biogenic CH₄ and N₂O emissions.^{7–10} However, they accounted for different emissions components inconsistently. For instance, U.S. EPA did not include upstream energy emissions,⁷ and only one inventory considered on-site, non-combustion CO₂ emissions.⁹ Seiple et al. catalogued U.S. facilities as part of their analyses to quantify the energy potential from wastewater sludge but did not estimate GHG emissions or energy consumption.¹¹

Therefore, we developed a novel approach to inventory wastewater emissions based on facility location and treatment processes, accounting for on-site, upstream, and downstream GHG sources. We estimated on-site emissions from treatment processes, on-site and upstream emissions from producing and using energy for facility operation and chemical production, and downstream emissions from offsite disposal of treated solid waste. We used this approach to generate a national emissions inventory of over 15,000 WWTPs across the contiguous United States. To capture uncertainty associated with various emission sources, we estimated potential emissions ranges using Monte Carlo simulations. Additionally, we conducted a global sensitivity analysis to identify key parameters influencing emissions, providing guidance for future data collection and decarbonization efforts. Our approach allows policymakers and engineers to analyze trade-offs inherent to different treatment technologies, better informing interventions for reducing wastewater treatment emissions. This method can be adapted as new measurement studies improve accuracy of emissions estimates and associated emissions factors.

For clarity, the following terminology is used in all text and figures: CH₄, N₂O, and CO₂ emissions are collectively described as "process emissions" and refer to the gases produced on-site during biological wastewater treatment processes, unless otherwise specified. CH₄ and N₂O produced from biosolids disposal are distinguished as "landfill CH₄" and "land application N₂O." Emissions

associated with natural gas include those from natural gas combusted on-site for heat (via boiler/dryer/incinerator) and chemical production, as well as upstream emissions from natural gas extraction and distribution. Electricity-related emissions are full fuel cycle, accounting for upstream and power plant emissions.

97

98

99

100

101

102

103

104

105

106

107

93

94

95

96

- Characterizing U.S. wastewater treatment facilities
- Our inventory includes 15,863 WWTPs in the contiguous United States. For each facility, we assigned a treatment train based on publicly available data reported by U.S. EPA. 12-15 We modeled 49 treatment configurations, representing the major combinations of processes used in the United States, each with liquids treatment, solids treatment, and additional optional processes (Figure 1). We refer to each configuration using the alphanumeric codes included in Figure 1, where letters indicate liquids treatment, numbers indicate solids treatment, and suffixes e and -p represent electricity generation and chemical phosphorous treatment, respectively (full details in Supplementary Methods Section 2.2.1.). We used energy and chemical requirements adapted from previous process models, 16 and determined process emissions based on treatment target and existing unit processes.

108 109

110

111

112

113

114

116

117

118

119

Figure 2a includes the total number of facilities and corresponding treated flow for each treatment configuration. Basic activated sludge (code A) accounts for 4,753 facilities (30% of total facilities) treating 23 billion m³·year⁻¹ (34% of annual flow). However, there are over 8,000 lagoons (aerobic, anaerobic, facultative, and unclassified) across the country, making them the most abundant technology despite only treating an estimated 7.8% of national flow. 32% of facilities use aerobic 115 digestion (code 3) for solids treatment and 19% use anaerobic digestion (codes 1 and 2). Treatment configurations with anaerobic digestion treat 64% of total flow while those with aerobic digestion only treat 17%, indicative of the large size of facilities with anaerobic digestion. Additionally, we identified 321 facilities recovering energy from biogas production, accounting for 10% of plants with anaerobic digestion. Facilities with energy recovery are large, with an average flow rate

- 120 (139,000 m³·day⁻¹, equivalent to 37 million gallons per day) greater than that of 98.6% of WWTPs
- included in this study.

122

- 123 Aside from lagoons, the most common treatment train is basic activated sludge coupled with
- aerobic digestion (code [*]A3), accounting for 22% (3,557) of all facilities and treating 10% of
- national flow (6.7 billion m³·year⁻¹). A lower proportion of all facilities (7.3%), basic activated
- sludge with anaerobic digestion ([*]A1 and [*]A1e) is the configuration that treats the most
- wastewater (17%). All nutrient transformation configurations combined (codes D, E, F, G) account
- for a small portion of total facilities (13%) but treat 39% of total national flow (27 billion
- $129 m^3 \cdot year^{-1}$).

130

- 131 Electricity and natural gas requirements
- We determined the electricity (kWh·m⁻³) and natural gas (MJ·m⁻³) needed for each treatment train
- 133 (Figure 2b) by expanding existing models¹⁶ to account for the full range of treatment
- 134 configurations in the United States. In this section and below, we report median results from our
- Monte Carlo simulation, with 5th and 95th percentiles reported subsequently in brackets. In Figure
- 2b, error bars (5th and 95th percentiles) reflect the national profile for electricity carbon intensity,
- highlighting the variability possible based on regional electricity mix (see Supplementary Methods
- Section 2.6.1.). The least energy intensive configurations include anaerobic / facultative lagoons
- 139 (L-n / L-f, 0.17 [0.14–0.21] kWh·m⁻³) and a trickling filter with anaerobic digestion (*C1e, 0.19
- 140 [0.083–0.29] kWh·m⁻³). These three configurations have no natural gas requirement. Conversely,
- the most intensive configurations use membrane bioreactors for biological nutrient removal (*D1e
- and *D3) which consume: 1.6 [0.83–2.3] kWh·m⁻³ electricity and 5.8 MJ·m⁻³ natural gas (*D1e,
- natural gas not subject to uncertainty for this configuration, Supplementary Table S24); and 1.7
- 144 [1.0-2.5] kWh·m⁻³ and 4.5 [3.9-5.2] MJ·m⁻³ (*D3). The electricity usage in these two
- 145 configurations is primarily for powering bioreactors, and the natural gas is for producing acetic
- acid used in nutrient removal.

- 148 With increased pumping and aeration for nitrification, the top electricity-consuming
- 149 configurations are those with biological nutrient removal for secondary treatment (codes D, E, F,
- 150 G). Within the top ten energy-consuming treatment configurations, process electricity accounts for

over 80% of total electricity consumption. Electricity for producing chemicals comprises a higher portion of emissions in biological phosphorous removal processes (codes D and G), where acetic acid comprises 9–22% of total electricity within baseline model inputs (mean: 15%). Consequently, regional grid largely determines electricity emissions rather than chemical manufacturing. Biological phosphorous removal requires the most natural gas, of which an average of 90% is used for acetic acid production, although this proportion decreases when solids are stabilized with incineration. Reducing natural gas dependency will require more sustainable chemical manufacturing and selection, as well as reduced reliance on incineration.

158159

160

151

152

153

154

155

156

157

- Carbon dioxide, methane, and nitrous oxide emissions
- Across different treatment trains, CH₄ and N₂O combined account for 73%–96% of median
- process emissions (CO₂, CH₄, N₂O from biological wastewater treatment), with CO₂ only
- exceeding 11% when organics removal (codes A, B, C) is coupled with aerobic digestion or
- incineration (codes 3, 4, 5, 6). The highest process emissions originate from facilities with
- nitrification (code E), anaerobic or facultative lagoons (L-f or L-n), and anaerobic digesters (codes
- 166 1 and 2) (Figure 2c). Nitrification with anaerobic digestion (*E1[e]) produces 0.69 [0.33–1.3] kg
- 167 CO_2 -eq·m⁻³, with 53% [24%–84%] from CH₄ and 40% [8.8%–72%] from N₂O. Anaerobic and
- 168 facultative lagoons produce 0.94 [0.13–2.3] kg CO₂-eq·m⁻³, 96% [70% –99%] of which is CH₄.
- However, each configuration's contribution to annual emissions depends on the abundance and
- 170 flow rate of facilities (Figure 2d). Nationwide, nitrifying facilities (code E) are the largest
- 171 contributors, generating an estimated 11 [6.9–17] million metric tonnes (MMT) CO₂-eq·year⁻¹,
- 172 followed by activated sludge facilities (code A), which contribute 7.5 [5.3–11] MMT CO₂-eq·year-
- 173 despite a much lower flow-normalized emission rate. All types of lagoons, in contrast, contribute
- 174 3.0 [1.5–4.9] MMT CO₂-eq·year⁻¹.

- Nationwide emissions by treatment configuration
- 177 Total emissions include process emissions, energy emissions, and downstream emissions from
- treated biosolids land application or landfill disposal. We did not include emissions from sewer
- systems, effluent discharge, and facility construction. To facilitate comparison between generic
- 180 configurations, we calculated emissions intensity (kg CO₂-eq·m⁻³) for each treatment train (Figure
- 2c and 2d) using the national profile for electricity carbon intensity (see Supplementary Methods

Section 2.6.1.). Note that total emissions associated with inventoried facilities use the carbon intensity of the plant's regional electricity supply (Table 1, and Figures 4 and 5). Highest total emissions are from configurations removing nutrients with membrane bioreactors: *D1e (1.8 [1.2– 2.6] kg CO_2 -eq·m⁻³) and *D3 (1.5 [0.85–2.4] kg CO_2 -eq·m⁻³). 59% [39%–77%] (*D1e) and 71% [45%–89%] (*D3) of emissions are from energy consumption, attributable to the electricity requirement of membrane systems and the natural gas needed for acetic acid production. The train with the next highest carbon intensity is *G1 (1.1 [0.73–1.7] kg CO₂-eq·m⁻³). Here, 60% [40%– 76%] of emissions are produced through biological treatment (CH₄, N₂O, CO₂) and 37% [22%– 56%] are from electricity and natural gas use. Across treatment trains, biosolids disposal at most accounts for 12% [4.6%–26%] of total emissions (*C4). Finally, by comparing median emission values from identical configurations with and without energy recovery through combined heat and power (CHP), we found that energy recovery only reduces total emissions by 5.2% to 13%. Understanding whether emissions are driven by treatment processes or electricity requirements will inform decarbonization strategies, particularly as the U.S. electricity generation mix continues to evolve. 17

We conducted a sensitivity analysis using Spearman's rank correlation to identify the following key drivers of emissions (p-value<0.05 and an absolute ρ-value>0.2) across treatment configurations: wastewater influent chemical oxygen demand (COD) and total nitrogen (TN), emissions factors for CH₄ and N₂O, electricity requirement, and grid carbon intensity (Figure 3). Across nearly all configurations, emissions are most sensitive to the CH₄ emission factor, even when anaerobic digestion is not present. In contrast, N₂O emissions factor and influent TN are primarily, though not exclusively, important for nutrient removal facilities. COD is a significant driver only in lagoon systems, for which fugitive CH₄ emissions are directly proportional (as reflected in CH₄ emissions factors). These findings underscore the importance of further refining emissions factors and influent wastewater variability through representative measurement campaigns and public datasets.

Nationally, wastewater treatment produces 47 [41–55] MMT CO₂-eq·year⁻¹ (Table 1) based on a national-scale Monte Carlo simulation. CH₄ dominates annual emissions (41% [31%–54%]), followed by N₂O (24% [16%–38%]) and electricity (23% [19%–27%]). Wider percentile ranges

- 213 for CH₄ and N₂O reflect higher uncertainty and greater variability in available emissions data.
- 214 Electricity emissions show lower uncertainty, reflecting less variation in estimates. While
- emissions from biosolids remain poorly characterized, our estimate of these emissions is only 2.2%
- 216 [1.1%–4.3%] of national emissions (1.1 [0.53–2.0] MMT CO₂-eq·year⁻¹). Emissions associated
- with natural gas production, distribution, and combustion are similarly low (1.4 [1.2–1.5] MMT
- 218 CO₂-eq·year⁻¹).

219

- 220 National distribution of GHG emissions from WWTP
- Figure 4 depicts the geographic distribution of emissions from selected treatment configurations:
- anaerobic digestion, *E1[e], F1[e], and lagoons (see Supplementary Figure S1 for similar plots for
- 223 each treatment train). Note that we only report baseline median values here for visual clarity.
- Emissions are highly distributed but cluster with major population centers, although nutrient
- removal configurations, particularly nitrification (*E1[e]), are more abundant in the eastern half
- of the country. The high density of anaerobic digestion facilities indicates that novel aerial methane
- measurement techniques may be promising for leak detection when used to survey large
- geographic areas. 18,19 While facilities with lagoons account for 9.6% of emissions in our inventory,
- these facilities are small, highly distributed, and largely of unknown operation, thus posing a
- potential challenge to mitigation efforts. In aggregate, we found that the largest 10% of emitters
- account for 82% of emissions (Figure 5a) and there is a strong linear relationship between total
- emissions and total flow (Figure 5b). The largest 10 emitters in the country (0.06% of facilities,
- treating 9.9% of national flow) generate 4.8 MMT CO₂-eq·year⁻¹ and disproportionately account
- for 11% of total emissions from all WWTPs (Figure 5c).

235

236

Discussion

- We estimate that emissions from wastewater treatment in the United States are 47 [41–55] MMT
- 238 CO₂-eq·year⁻¹, with CH₄ and N₂O accounting for 66% [51%–86%] of emissions. Compared to
- previous works (Supplementary Table 1), our approach integrates a greater number of emissions
- sources, including electricity, CO₂ (fossil-origin), and biosolids disposal, and allows for a side-by-
- side comparison across key emissions types. Our estimates of GHG intensity for specific treatment
- 242 trains range from 0.33 [0.19–0.74] kg CO_2 -eq·m⁻³ (*C6) to 1.8 [1.2–2.6] kg CO_2 -eq·m⁻³ (*D1e),
- aligning with models by U.S. EPA (0.5-1.8 kg CO₂-eq·m⁻³), although the relative contributions of

different components differ.²⁰ Largely due to a higher number of identified anaerobic digestion facilities, our national estimate of CH₄ emission (19 [15–25] MMT CO₂-eq·year) is approximately twice that of Song et al., who estimated 9.7 MMT CO₂-eq·year by drawing on one of the four source datasets used in this study (see Methods for additional details).³ Our nationwide estimate for N₂O (12 [7.9–17] MMT CO₂-eq·year) is similar to values reported by Song et al. (11.6 MMT CO₂-eq·year), calculated using the same underlying emission factor data which we grouped by treatment objected, whereas Song et al organized by bioreactor type.⁵

251252

253

254

255

256

257

258

259

260

261

262

263

264

265

266

267

244245

246

247

248

249

250

Methane emissions are the largest single contributor to annual emissions in our inventory. We found 85% [74%–92%] (16 [12–21] MMT·year-1) of CH₄ is produced by treatment configurations with anaerobic digestion (code 1 or 2). Anaerobic digestion produces energy on-site in the form of biogas, while reducing the spatial footprint of WWTPs and providing additional opportunities for resource recovery.²¹ However, fugitive CH₄ emissions currently outweigh climate benefits gained from renewable biogas. On a volumetric basis, energy recovery (code suffix e) only reduces the median emissions by an average of 0.061 kg CO₂-eq·m⁻³. In contrast, the average of median emissions from treatment configurations with anaerobic digesters is 0.34 kg CO₂-eq·m⁻³, or 0.30 kg CO₂-eq·m⁻³ higher than those with aerobic digestion. While our analysis uses data from the United States, these findings have broader global implications as concerns about GHG emissions from similar treatment trains are global. Recent studies in Europe have identified key sources of methane leaks in anaerobic digestion facilities, including non-gastight digestate storage, ²² biomass storage tanks, and pressure relief valves.²³ Additionally, minor technical fixes to existing infrastructure have been demonstrated to reduce methane emissions from biogas facilities by up to 46%,²³ highlighting the potential impact of leak detection and repair programs and the importance of immediately adopting such a program globally.

268

269

270

271

272

273

274

We developed N₂O emission factors adapted from those compiled by Song et al. 2024, distinguishing based on treatment objectives (organics removal, nitrification, denitrification).⁵ However, 79% of the underlying measurement data comes from denitrifying systems.²⁴ Additional measurements are needed from conventional activated sludge and nitrifying systems. Given the high spatial and temporal variability in N₂O emissions from wastewater treatment,⁶ improving the characterization of N₂O production is essential. Additionally, current N₂O mitigation efforts use

aeration, feed, and process optimization, ²⁵ which are not captured in existing facility-level data.

276 Understanding current operation strategies and their impact on emissions should be a focus of

277 future research.

278

279

280

281

282

283

284

285

The relative importance of electricity generation (23% [19%–27%] of total emissions) will decrease with grid decarbonization. Biosolids disposal through landfilling or land application only contribute a small portion of total emissions (2.2% [1.1%–4.3%]), but CH₄ and N₂O from biosolids are poorly studied. We used IPCC's emission factor for managed soils, which has a high uncertainty (0.002–0.018 kg N₂O-N·kg N⁻¹)²⁷ but align with the small number of recent studies where biosolids were used as an agricultural amendment. Finally, our model did not account for CH₄ produced through different practices for biosolids dewatering and on-site storage, which

also contribute to fugitive CH₄ emissions.³¹

287288

289

290

291

292

293

294

295

296

297

298

299

300

301

302

303

U.S. EPA provides, to the best of our knowledge, the only other national level inventory of wastewater treatment emissions in the United States. The two inventories differ in that we included electricity-associated emissions in order to enable a side-by-side comparison of key emissions sources, reflecting the wastewater industry's historic prioritization of achieving net-zero energy use. Also differing from EPA, we did not include CH₄ and N₂O effluent discharge emissions because we focused on emissions that can inform decarbonization efforts. Comparing on-site emissions, our N₂O estimate (12 [7.9–17] MMT·year⁻¹) is comparable to the EPA's value (16.3 MMT·year⁻¹), although our CH₄ emissions estimate (19 [15–25] MMT·year⁻¹) are threefold greater than their estimate (5.6 MMT·year⁻¹). The relative contributions of the two gases also differ, with CH₄ contributing 63% [51%–73%] (19 [15–25] MMT·year⁻¹) of onsite CH₄ and N₂O emissions in our inventory but only 26% (5.6 MMT) in the EPA inventory. This difference is meaningful, as the relative importance of each gas informs potential mitigation measures. CH₄ and N₂O emissions are produced by different wastewater treatment processes via distinct microbial pathways, which are impacted by local climate (e.g. temperature and seasonal changes to operation^{6,32}). Additionally, atmospheric lifespan and global warming potential will impact the priority of mitigation strategies for these two gases.

There are several limitations to this work. Our analysis relies on data reported through U.S. EPA's Clean Watersheds Needs Surveys (CWNS), which did not require reporting unit process data in the most recent 2022 survey. As a result, 75% of our inventory relies on unit process data from 2012, although we used supplemental datasets and manual verification to update the data where possible (Figure 6; Supplementary Methods Section 2.2.2.). Furthermore, our results indicate a strong linear relationship between cumulative national flow rate and emissions (Figure 5B). However, most recent national flow data from the CWNS does not distinguish between observed and design flow at facilities, meaning we likely overestimated the volume of wastewater treated. Additionally, we used uniform average influent concentrations for all facilities, and did not account for variations in effluent requirements based on local regulations. Given their importance in estimating GHG emissions, future CWNS surveys should require data reporting on: treatment processes; biogas production and utilization; observed flow; influent and effluent water quality.

There are also several opportunities for further refining in future inventories, particularly with expanded data collection. We made the simplifying assumption that, within a particular treatment plant configuration, energy consumption is directly proportional to flow rate, despite the potential for increased efficiency at larger sizes.³³ Additionally, the energy requirements for lagoons may not reflect current designs and operation.³⁴ Future inventories and resulting mitigation efforts will also benefit from improved CH₄ and N₂O emissions factors, which will require direct measurement studies across a range of representative facilities, climates, and time periods. Additionally, for lagoons, we used current IPCC assumptions that aerobic lagoons do not produce CH₄ and that anaerobic lagoons do not produce N₂O. However, given lack of fully uniform mixing, and the many connections between CH₄ and nitrogen microbial metabolism,³⁵ it is likely both gases are produced across all lagoon categories.

Our analysis only considered climate impact, but facility-level decision making requires more comprehensive environmental assessment. For example, trickling filter configurations are among those with the lowest total emissions. However, they have large spatial requirements and require adequate hydraulic head differences across the plant to limit pumping requirements. Additionally, because anaerobic digesters are the largest source of on-site emissions, facilities using incineration (codes 5 and 6) compare favorably because they have lower on-site emissions. However,

incineration requires adequate pollution control measures to prevent release of particulates, heavy metals, and volatile organic compounds, considerations that we did not capture.

338

339

340

341

342

343

344

345

346

347

348

349

350

336

337

Wastewater treatment is a growing sector inextricably linked to public and environmental health, and understanding its current climate change impact is critical for decarbonization efforts. This analysis provides a comprehensive inventory of over 15,000 WWTPs across the contiguous United States, including treatment configuration, energy requirements, and emissions estimates. While confined to the United States, a similar approach can be adopted elsewhere. In less data-rich contexts, the archetypical treatment configurations reported here can be selected based on best fit with available information, or serve as a guide for future data collection. In the United States, we identify on-site emissions of CH₄ and N₂O as priorities for climate change mitigation efforts, expanding the historical focus of the wastewater sector beyond energy-associated emissions. Additionally, data generated from this work can be used to determine the impact of specific policy levers on overall emissions, and to analyze the effects of adopting novel resource recovery and emissions reduction technologies.

351

352

Methods

- 353 Facilities inventory development
- We integrated multiple national datasets to compile an inventory of wastewater treatment facilities
- in the United States, their energy requirements, and greenhouse gas emissions (Figure 6). Our
- inventory consists of 15,863 wastewater treatment plants that reported non-zero flow to the U.S.
- 357 Environmental Protection Agency (EPA) in 2022.¹⁵ Throughout this manuscript, wastewater
- 358 treatment plants (WWTPs) refer to facilities owned and/or operated by municipalities or other
- public entities, including treatment plants and lagoons. We determined the treatment train for each
- 360 facility in our inventory, defined as a common set of unit operations designed to reduce wastewater
- pollution between the influent and effluent of the plant. We used unit processes reported across
- the aggregated 2004, 2008, 2012, and 2022 Clean Watersheds Needs Surveys (CWNS), 12-15
- 363 supplemented with additional publicly available data that provided more granular or recent
- information: EPA's Lagoon Inventory Dataset, ³⁶ Water Environment Federation's (WEF) Water
- Resource Recovery Facilities Biogas Database,³⁷ and U.S. Department of Energy's (DOE)
- 366 Combined Heat and Power Installation Database.³⁸

Using reported unit process data, we assigned each facility one or more treatment trains, abbreviated using alphanumeric codes, based on those previously defined by Tarallo et al., 2015¹⁶ but with modifications to reflect additional possible combinations of liquids and solids treatment processes (full details are provided in Supplementary Methods Section 2.2.2.) Each treatment train is a unique combination of unit processes depicted in Figure 1, and each configuration includes liquids and solids treatment processes, with optional primary treatment, chemical phosphorous removal, and CHP for energy recovery. Liquids (or secondary) treatment technologies include activated sludge configurations, trickling filters, and membrane bioreactors. Solids treatment options include aerobic and anaerobic digestion, lime stabilization, and two incineration methods (see Supplementary Tables S2 and S3 for additional details). With available unit process data, we assigned treatment trains to 10,962 facilities, approximately 69% of the national fleet.

Where unit process data was incomplete or unavailable, we assigned treatment trains based on facilities of a similar size and geographic location. For the 1,991 facilities with only partial unit process data available (i.e., plants that provide information on secondary treatment or solids management but not both), we assigned treatment trains based on the most common treatment train of the same plant size and EPA region, considering key unit processes present (activated sludge, biological nutrient removal, aerobic/anaerobic digesters, lime stabilization, incineration, and trickling filters). For the remaining facilities with either insufficient partial data or fully absent data (2,910 facilities), we assigned a treatment train based on the most common treatment train of the same plant size and EPA region. Supplementary Figure S2 and Table S7 provide a size breakdown and additional details on facilities with missing data.

- Facility-level emissions associated with energy
- For all treatment trains in the national inventory, we calculated electricity and natural gas consumption, as well as on-site electricity generation from biogas utilization. We used energy calculations from the results of process models in GPS-XTM reported by Tarallo et al., 2015.¹⁶ Because our study includes treatment trains beyond those reported by Tarallo et al., we used mass and heat balances for unit processes to determine energy requirements for treatment trains that were not modeled previously (details in Supplementary Methods Section 2.3.1.).¹⁶

398399

400

401

402

403

404

405

406

407

408

To estimate the GHG emissions associated with electricity consumption, we assigned each facility a regional emissions factor (kg CO₂-eq·kWh⁻¹). Specifically, we used existing model results that simulate energy mix across 134 simulated sub-regions of the United States, reflecting state-level and utility boundaries, and existing variation to maintain load-interchange-generation balance.³⁹ We calculated electricity emissions factor (kg CO₂-eq·kWh⁻¹) for each geographic region using the corresponding total emissions (kg CO₂-eq) divided by the net power generated, as reported in NREL Standard Scenarios Cambium data for 2020.^{17,40} We used data from the mid-case scenario based on central parameter values, including future electricity consumption, fuel costs, and technology selection. We also calculated full fuel cycle GHG emissions for electricity and natural gas using the Greenhouse gases, Regulated Emissions, and Energy use in Technologies (GREET) model (Supplementary Table S18).⁴¹

409410

411

412

413

414

415

416

417

418

419

420

421

422

423

424

425

426

427

428

Carbon dioxide, methane, and nitrous oxide

We estimated total CO₂, CH₄, and N₂O from biological treatment processes, referred to here as "process emissions" for brevity. For CO₂, we assumed a baseline of 11.9% of influent carbon is of fossil origin, and 53.5% of influent COD (508 mg·L⁻¹) is released as CO₂ during biological treatment. ⁴² To determine CH₄ production, we used emissions factors based on the presence of key unit processes, and for N₂O production we used emissions factors based on treatment targets. Namely, anaerobic processes (anaerobic digestors and lagoons) emit CH₄, and nutrient removal processes emit N₂O. For CH₄, we used data reported by Song et al. 2023 to assign emissions based on whether a facility contains an anaerobic digester.³ For facilities with anaerobic or facultative lagoons, we used emission factors reported by IPCC. 43 For N₂O, we developed emissions factors using data compiled in Song et al., 2024 to determine median value of literature-reported values categorized by treatment objective: organics removal, nitrification, or full denitrification (Supplementary Table S20).⁵ N₂O emissions factors are a function of influent total nitrogen, and we assumed a range typical of domestic wastewater (23–69 mg/L).⁴⁴ As with CH₄, we used N₂O emissions factors reported by IPCC for aerobic, anaerobic, and facultative lagoons.⁴³ For uncategorized lagoons, we calculated both CH₄ and N₂O emissions factors using a flow weighted average of the three other types of lagoons in the contiguous United States. For specific emissions factors and additional details, see Supplementary Methods Section 2.4.

429 430 Biosolids associated emissions 431 To estimate biosolids production, we combined reported production rates, where data was 432 available, with mass flow calculations where data was unavailable. For 2,124 facilities, we used EPA's Biosolids Biennial Report for 2020-2021, 45 which documents the volume of biosolids 433 434 produced, and their ultimate disposal through incineration, landfilling, or land application. For the 435 remaining facilities, we calculated sludge production based on facility flow rate, as developed elsewhere and described in Supplementary Methods Section 2.5.1.¹¹ For land applied biosolids, 436 we used the IPCC N₂O emissions factor for organic soil amendments.²⁷ For landfills, we used U.S. 437 438 EPA's Landfill Gas Emissions Model (LandGEM) to calculate the CH₄ emission factor for 439 municipal solid waste landfills. 46 Full details are included in Supplementary Methods Section 440 2.5.2. 441 442 *Uncertainty and sensitivity analyses* 443 For all treatment trains, we conducted Monte Carlo simulations (N = 10,000) for 19 input 444 parameters subject to uncertainty. We selected all uncertainty ranges and distributions (Supplementary Table S24) using the tiered selection criteria previously developed, ^{47,48} with minor 445 modifications described in Supplementary Methods Section 2.6. Sensitivity analyses evaluated the 446 447 relative impact of all input parameters on total per-volume GHG emissions for each treatment 448 train, using Spearman's rank order correlation coefficients. In this analysis, we defined key drivers 449 of uncertainty as those with a p-value<0.05 and an absolute ρ-value>0.2. 450 451 For national-level results, we also evaluated uncertainty around two additional inputs: local 452 electricity carbon intensity and facility biosolids production. We determined baseline facility-453 specific carbon intensity using 134 geographic regions reported in NREL Standard Scenarios Cambium data for 2020, 17,40 and determined emissions factors for each electricity production 454 455 pathway using the GREET model.⁴¹ For local electricity carbon intensity, we assumed a uniform 456 distribution with 80% and 120% of the baseline value as the lower and upper bound, respectively. 457 For biosolids, we calculated the baseline facility-level mass flow rate and determined disposal 458 pathways as described above and in the Supplementary Methods Section 2.5. Next, we used 459 facility-level data to calculate the national total biosolids mass flow to landfills and land

- application. For our uncertainty analysis, we assumed a uniform distribution with 80% and 120%
 of the national total biosolids mass flow rate as the lower and upper bounds, respectively. Full
- details are provided in Supplementary Methods Sections 2.5 and 2.6.
- Due to the high number of facilities in this study, we only report median values for all facility-
- level results (Figures 4, 5 and Supplementary File C). Specifically, for each WWTP, we used
- 466 median emissions factors to estimate emissions from individual processes (e.g., biological CH₄
- emissions, upstream electricity emissions), and summed across all processes at a given facility to
- determine total emissions.

470 **Data Availability**

463

469

484

485

487

488

- The data used in the analysis and to generate figures are publicly available, with the exception of
- underlying data from process models in Tarallo et al. 2015¹⁶ and raw data files from Song et al.,
- 473 2023³ and Song at al., 2024,⁵ the three of which were shared by the authors upon request. Data
- 474 from the Clean Watersheds Needs Survey is available through the U.S. EPA
- 475 (https://www.epa.gov/cwns). Data on electricity generation from biogas is available through the
- 476 U.S. Department of Energy's Combined Heat and Power Installation Database
- 477 (https://doe.icfwebservices.com/chp) and the Water Environment Federation's Water Resource
- 478 Recovery Facilities Biogas Database (https://www.resourcerecoverydata.org/). Data used to
- 479 identify lagoons is available through the U.S. EPA (https://www.epa.gov/small-and-rural-
- 480 wastewater-systems/lagoon-wastewater-treatment-systems#dataset). Electricity grid data is
- available through the National Renewable Energy Laboratory (https://scenarioviewer.nrel.gov/).
- 482 The datasets generated through this work are publicly available at:
- 483 <u>https://doi.org/10.5281/zenodo.15857432.</u>

Code Availability

All code supporting this study is available at https://github.com/jiananf2/US WWTP GHG.

Acknowledgements

- 489 This study was funded by the U.S. Department of Energy Industrial Technologies Office (all
- 490 authors). J.F. also received support from the Loh Kwan Chen fellowship at the University of

- 491 Illinois Urbana-Champaign. We thank Sabyasachi Das for contributing to the early stages of this
- analysis. Heroda Abera conducted manual checks for selected wastewater treatment facilities.
- 493 Maram Canawati provided administrative support. We also thank Trung Le for input on anaerobic
- 494 digester emissions, Saumitra Rai for discussion on sludge incineration, and Professor Khaled
- 495 Abdel-Ghaffar for support in data acquisition from libraries at University of California, Davis.
- 496
- This work was authored in part by the National Renewable Energy Laboratory, operated by
- 498 Alliance for Sustainable Energy, LLC, for the U.S. Department of Energy (DOE) under Contract
- No. DE-AC36-08GO28308 and the Lawrence Berkeley National Laboratory under Contract No.
- 500 DE-AC02-05CH11231. The views expressed in the article do not necessarily represent the views
- of the DOE or the U.S. Government. The U.S. Government retains and the publisher, by accepting
- the article for publication, acknowledges that the U.S. Government retains a nonexclusive, paid-
- 503 up, irrevocable, worldwide license to publish or reproduce the published form of this work, or
- allow others to do so, for U.S. Government purposes.

505

506

Author contributions

- 507 Conceptualization was performed by J.S.D., J.B.D, S.H.E., J.F., J.M., M.M.B., M.A.,
- Methodology was developed by J.S.D., S.H.E., J.F., J.B.D., J.S.G., A.R.H., M.M.B., M.A., J.M.
- Validation was conducted by J.F., S.H.E., A.R.H., C.P. Data Curation was performed by J.F., A.H.,
- 510 C.P., M.B. Formal analysis and software development was performed by J.F., A.R.H., S.H.E.,
- J.S.D., P.Z., M.B., M.A., Investigation was performed by S.H.E., J.F., J.S.G., M.B., M.A.
- Visualization was done by J.F. and S.H.E. Original draft was written by S.H.E., J.B.D., J.F.,
- 513 A.R.H., and M.A. Writing reviewing and editing was performed by all authors. Funding
- acquisition was conducted by J.S.D., J.B.D., and J.M. Supervision was performed by J.S.D.,
- 515 J.B.D., J.S.G., and S.H.E.

516517

Competing Interests

518 The authors declare no competing interests.

519 Tables

520 521

522

523

524

Table 1. Annual emissions from wastewater treatment in the United States, reported as median values followed by 5^{th} to 95^{th} percentile values in brackets

Emissions Type		Annual Emissions (MMT CO ₂ -eq·year ⁻¹)	Percent of total emissions	
Process emissions	Methane	19 [15–25]	41% [31%–54%]	
(produced through	Nitrous oxide	12 [7.9–17]	24% [16%–38%]	
biological treatment)	Carbon dioxide	2.8 [2.2–3.5]	5.9% [4.5%–7.7%]	
Energy emissions	Electricity*	11 [9.8–12]	23% [19%–27%]	
	Natural gas (combustion for on- site processes & chemical production)	1.1 [1.0–1.2]	2.4% [2.0%–2.8%]	
	Natural gas production & distribution	0.25 [0.23–0.27]	0.53% [0.45%–0.63%]	
Downstream emissions	Landfill methane	0.35 [0.28–0.43]	0.74% [0.57%–0.95%]	
from biosolid disposal	Land application nitrous oxide	0.73 [0.2–1.6]	1.5% [0.40%–3.5%]	
Total		47 [41–55]	100%	

^{*} Electricity estimates in the national inventory are based on local balancing area carbon intensities.

525 Figures

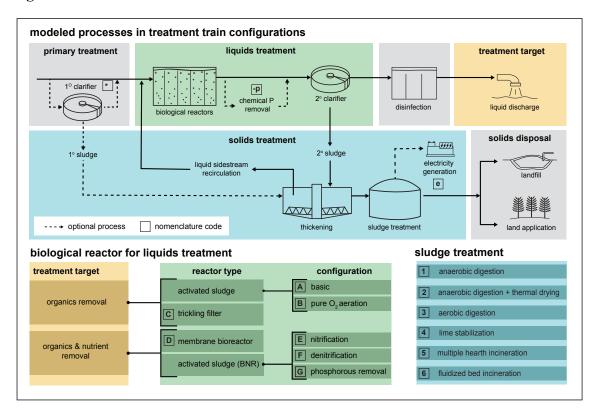


Figure 1. Overview of treatment configurations. Top panel: modeled unit processes for each treatment train, with all major energy consuming processes. Variations in treatment train are determined based on the biological reactor and its configuration (bottom left, in green) and solids management (bottom right). The following processes may also be included or excluded (indicated with the dashed lines): primary treatment, chemical phosphorous removal, and power generation. Naming convention for each treatment train is based on the combination of biological reactor and solids management. The * prefix is added to indicate the presence of primary treatment, and the suffixes -p and e indicate the presence of chemical phosphorous removal and power generation, respectively. All nomenclature codes are depicted in boxes next to the relevant process in this figure. Detailed descriptions of each treatment train can be found in Supplementary Tables S2 and S3.

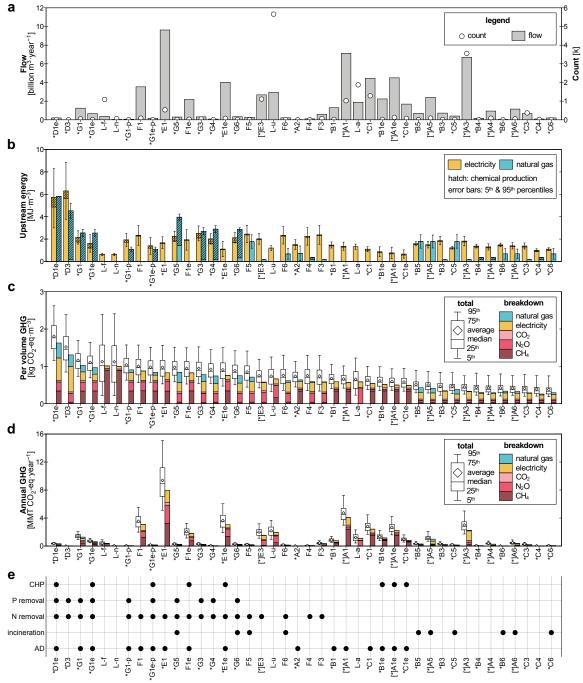


Figure 2. Treatment train distribution and associated emissions. a, count and flow; b, electricity and natural gas consumption intensity; c, greenhouse gas (GHG) emissions intensity; d, annual GHG emissions; e, key unit processes and treatment targets of treatment trains. Identical configurations with and without primary treatment are combined using brackets ([*]). Treatment train codes are defined in Figure 1, trains beginning with L- indicate lagoons (L-a: aerobic; L-n: anaerobic; L-f: facultative; L-u: uncategorized, calculated as the flow-weighted average of aerobic and anaerobic/facultative lagoons). In b, hatches indicate energy used for chemical production (methanol, acetic acid, hypochlorite, and lime). In c and d, the electricity emissions reflect national average carbon intensity of electricity production. In b, c, and d, error bars reflect results of 10,000 Monte Carlo simulations, also summarized in box plots (whiskers indicate 5th and 95th percentiles, boxes indicate the 25th to 75th percentiles, midlines indicate the 50th

percentiles, and diamonds represent the mean). In b, c, and d, colors indicate different emission types: yellow indicates upstream emissions associated with electricity usage; blue indicates emissions associated with natural gas extraction, transportation, and on-site combustion; red indicates on-site GHG emissions, with shade indicating gas type: CH_4 (dark), N_2O (medium), CO_2 (light). Emissions associated with biosolids handling have limited contribution to the total emission and are not included in breakdown bars for figure clarity, but values remain included in box plot results. Sum of the median value of each emission type (bar plots) in c and d tends to be lower than median of total emissions (box plots) due to right-skewed underlying distributions, where extremely high values can disproportionately influence aggregate median. In e, circles indicate the presence of specified unit processes or treatment targets. Abbreviations: CHP: combined heat and power; AD: anaerobic digestion; P removal: phosphorous removal; N removal: nitrogen removal.



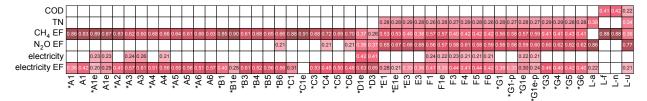


Figure 3. Sensitivity of emissions estimates to key input parameters. Values represent Spearman's ρ -value for significant parameters (ρ -value<0.05 and absolute ρ -value>0.2), indicating their influence on per volume emission for each treatment configuration. An absolute ρ -value closer to 1 (darker red) indicates the corresponding parameter has stronger influence on the emission result. Abbreviations: COD = chemical oxygen demand in plant influent, TN = total nitrogen in plant influent, EF = configuration.

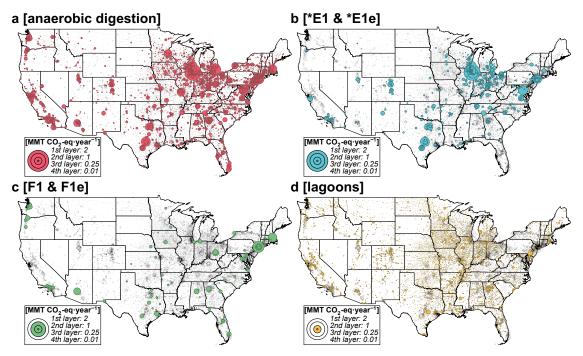


Figure 4. Geographic distribution of emissions from selected treatment processes and trains. a, all facilities with anaerobic digestion (codes 1 and 2), b, *E1 and *E1e, c, F1 and F1e, and d, all lagoons (aerobic, anaerobic, facultative and unclassified). Legends from inner to outer rings represent annual

emission of 0.01, 0.25, 1, and 2 MMT CO_2 -eq, respectively. Non-shaded legend indicates no wastewater treatment plant has emission in the corresponding level.

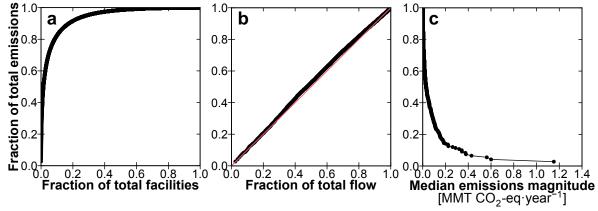


Figure 5. **Distribution of total emissions in the United States**. In each plot, individual points represent a single facility, and the y-axis represents the fraction of total emissions. The x-axes represent: a, fraction of total facilities, with facilities sorted from highest to lowest emissions, b, fraction of total flow when facilities are sorted by flow rate from largest to smallest, and c, magnitude of emissions from a single facility, with facilities sorted from lowest to highest emissions. The red line in b represents parity of x- and y-axes values (y=x).

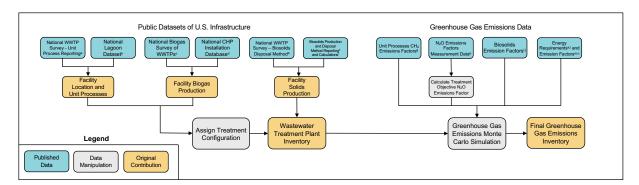


Figure 6. Dataset integration workflow for facility inventory, energy consumption and GHG emissions. References listed in the flow chart are: "Clean Watersheds Needs Surveys (2004, 2008, 2012, 2022)^{12–15} bU.S. EPA's inventory of lagoons serving as the primary method of wastewater treatment at a given facility, ³⁶ Water Environment Federation survey of wastewater treatment plants with biogas production, ³⁷ dU.S. Department of Energy's survey of facilities with combined heat and power, ³⁸ EPA annual report on biosolids production and disposal methods, ⁴⁵ Biosolid calculations outlined in Seiple et al., 2017, ¹¹ Song et al 2023 methane emissions factors from wastewater treatment plants, ³ Song et al 2024 nitrous oxide emissions factors for wastewater treatment plants, ⁵ IPCC emissions factors for land applied biosolids, ²⁴ and ^jU.S. EPA's LandGEM model for estimating methane emissions from landfills ⁴⁶ Tarallo et al, 2015 modelled energy requirements for treatment trains, ¹⁶ U.S. EPA estimate of lagoon energy requirements, ³⁴ GREET model, ⁴¹ IPCC emissions factors for lagoons².

596 References

- 1. Song, C., Zhu, J.-J., Yuan, Z., Van Loosdrecht, M. C. M. & Ren, Z. J. Defining and
- achieving net-zero emissions in the wastewater sector. *Nat. Water* **2**, 927–935 (2024).
- 599 2. Industry. in Climate Change 2022 Mitigation of Climate Change (ed. Intergovernmental
- Panel On Climate Change (IPCC)) 1161–1244 (Cambridge University Press, 2023).
- doi:10.1017/9781009157926.013.
- 3. Song, C. et al. Methane Emissions from Municipal Wastewater Collection and Treatment
- 603 Systems. *Environ. Sci. Technol.* **57**, 2248–2261 (2023).
- 4. Moore, D. P. et al. Underestimation of Sector-Wide Methane Emissions from United States
- Wastewater Treatment. *Environ. Sci. Technol.* **57**, 4082–4090 (2023).
- 5. Song, C. et al. Oversimplification and misestimation of nitrous oxide emissions from
- wastewater treatment plants. *Nat. Sustain.* (2024) doi:10.1038/s41893-024-01420-9.
- 608 6. Gruber, W. et al. Estimation of countrywide N2O emissions from wastewater treatment in
- Switzerland using long-term monitoring data. Water Res. X 13, 100122 (2021).
- 7. U.S. Environmental Protection Agency. *Inventory of U.S. Greenhouse Gas Emissions and*
- Sinks 1990-2022. https://www.epa.gov/ghgemissions/inventory-us-greenhouse-gas-
- 612 emissions-and-sinks-1990-2022 (2023).
- 8. Parravicini, V. Evaluation of greenhouse gas emissions from the European urban wastewater
- sector, and options for their reduction. Sci. Total Environ. (2022).
- 615 9. Wang, D. et al. Greenhouse gas emissions from municipal wastewater treatment facilities in
- 616 China from 2006 to 2019. *Sci. Data* **9**, 317 (2022).
- 10. Chen, W., Zhang, Q., Hu, L., Geng, Y. & Liu, C. Understanding the greenhouse gas
- emissions from China's wastewater treatment plants: Based on life cycle assessment coupled
- with statistical data. *Ecotoxicol. Environ. Saf.* **259**, 115007 (2023).

- 620 11. Seiple, T. E., Coleman, A. M. & Skaggs, R. L. Municipal wastewater sludge as a sustainable
- bioresource in the United States. J. Environ. Manage. 197, 673–680 (2017).
- 622 12. U.S. Environmental Protection Agency. Clean Watersheds Needs Survey 2004. (2008).
- 13. U.S. Environmental Protection Agency. Clean Watersheds Needs Survey 2008. (2010).
- 14. U.S. Environmental Protection Agency. Clean Watersheds Needs Survey 2012. (2016).
- 625 15. U.S. Environmental Protection Agency. Clean Watersheds Needs Survey 2022. (2024).
- 626 16. Tarallo, S., Shaw, A., Kohl, P. & Eschborn, R. A Guide to Net-Zero Energy Solutions for
- Water Resource Recovery Facilities. https://iwaponline.com/ebooks/book/293/ (2015).
- 628 17. Gagnon, P., Frazier, W., Hale, E. & Cole, W. Cambium data for 2020 Standard Scenarios.
- National Renewable Energy Laboratory (2020).
- 18. Bell, C. et al. Single-blind determination of methane detection limits and quantification
- accuracy using aircraft-based LiDAR. Elem. Sci. Anthr. 10, 00080 (2022).
- 632 19. El Abbadi, S. H. et al. Technological Maturity of Aircraft-Based Methane Sensing for
- Greenhouse Gas Mitigation. *Environ. Sci. Technol.* acs.est.4c02439 (2024)
- 634 doi:10.1021/acs.est.4c02439.
- 635 20. U.S. Environmental Protection Agency. Life Cycle and Cost Assessments of Nutrient
- Removal Technologies in Wastewater Treatment Plants.
- https://www.epa.gov/system/files/documents/2021-08/life-cycle-nutrient-removal.pdf
- 638 (2021).
- 639 21. El Abbadi, S. H. & Criddle, C. S. Engineering the Dark Food Chain. *Environ. Sci. Technol.*
- **53**, 2273–2287 (2019).

- 641 22. Wechselberger, V. et al. Assessment of whole-site methane emissions from anaerobic
- digestion plants: Towards establishing emission factors for various plant configurations.
- 643 *Waste Manag.* **191**, 253–263 (2025).
- 644 23. Michael Fredenslund, A., Gudmundsson, E., Maria Falk, J. & Scheutz, C. The Danish
- national effort to minimise methane emissions from biogas plants. *Waste Manag.* **157**, 321–
- 646 329 (2023).
- 647 24. De Haas, D. & Andrews, J. Nitrous oxide emissions from wastewater treatment Revisiting
- the IPCC 2019 refinement guidelines. *Environ. Chall.* **8**, 100557 (2022).
- 649 25. Duan, H. et al. Insights into Nitrous Oxide Mitigation Strategies in Wastewater Treatment
- and Challenges for Wider Implementation. *Environ. Sci. Technol.* **55**, 7208–7224 (2021).
- 26. Li, X., Song, C., El Abbadi, S. H., Stokes-Draut, J. R. & Ren, Z. J. The Impact of Clean Grid
- Transition on Wastewater Sector Greenhouse Gas Emissions. *Environ. Sci. Technol. Lett.* 12,
- 653 144–150 (2025).
- 654 27. Hergoualc'h, K. et al. N2O Emissions From Managed Soils, And CO2 Emissions from Lime
- and Urea Application. in 2019 Refinement to the 2006 IPCC Guidelines for National
- 656 Greenhouse Gas Inventories (Intergovernmental Panel on Climate Change, Switzerland,
- 657 2019).
- 28. Obi-Njoku, O. et al. A comparison of Tier 1, 2, and 3 methods for quantifying nitrous oxide
- emissions from soils amended with biosolids. Sci. Total Environ. 915, 169639 (2024).
- 29. Obi-Njoku, O. et al. Greenhouse gas emissions following biosolids application to farmland:
- Estimates from the DeNitrification and DeComposition model. Sci. Total Environ. 823,
- 662 153695 (2022).

- 30. Roman-Perez, C. C., Hernandez-Ramirez, G., Kryzanowski, L., Puurveen, D. & Lohstraeter,
- G. Greenhouse gas emissions, nitrogen dynamics and barley productivity as impacted by
- biosolids applications. *Agric. Ecosyst. Environ.* **320**, 107577 (2021).
- 31. Oshita, K. et al. Methane and nitrous oxide emissions following anaerobic digestion of
- sludge in Japanese sewage treatment facilities. *Bioresour. Technol.* **171**, 175–181 (2014).
- 32. Roothans, N. et al. Long-term multi-meta-omics resolves the ecophysiological controls of
- seasonal N2O emissions during wastewater treatment. *Nat. Water* (2025)
- doi:10.1038/s44221-025-00430-x.
- 33. Palgrave, R. Centrifugal pump basics; part 1. in *Troubleshooting Centrifugal Pumps and*
- *their Systems* 13–60 (Elsevier, 2020). doi:10.1016/B978-0-08-102503-1.00003-7.
- 34. U.S. Environmental Protection Agency. *Principles of Design and Operations of Wastewater*
- 674 Treatment Pond Systems for Plant Operators, Engineers, and Managers.
- https://www.epa.gov/sites/default/files/2014-09/documents/lagoon-pond-treatment-2011.pdf
- 676 (2011).
- 35. Stein, L. Y. & Lidstrom, M. E. Greenhouse gas mitigation requires caution. *Science* **384**,
- 678 1068–1069 (2024).
- 679 36. U.S. Environmental Protection Agency. Lagoon Inventory Dataset. (2022).
- 680 37. Water Environment Federation (WEF). Water Resource Recovery Facilities Biogas Data.
- 681 (2024).
- 38. U.S. Department of Energy (DOE). U.S. Department of Energy Combined Heat and Power
- 683 Installation Database. (2024).
- 684 39. NREL. The Role of Large Balancing Areas in Integrating Solar Generation.
- https://www.nrel.gov/docs/fy11osti/50059.pdf (2011).

- 40. Cole, W. et al. Regional Energy Deployment System Model 2.0 (ReEDS 2.0). (2021).
- 41. Argonne National Laboratory. GREET Model.
- 42. Law, Y., Jacobsen, G. E., Smith, A. M., Yuan, Z. & Lant, P. Fossil organic carbon in
- wastewater and its fate in treatment plants. *Water Res.* 47, 5270–5281 (2013).
- 690 43. Bartram, D. et al. Wastewater Treatment and Discharge. in 2019 Refinement to the 2006
- 691 IPCC Guidelines for National Greenhouse Gas Inventories (Intergovernmental Panel on
- 692 Climate Change, 2019).
- 693 44. Tchobanoglous, G. et al. Chapter 3: Wastewater Flowrates and Constituent Loadings. in
- Wastewater Engineering: Treatment and Resource Recovery (McGraw-Hill Education,
- 695 2014).
- 696 45. U.S. Environmental Protection Agency. Biosolids Biennial Report No. 9 (Reporting Period
- 697 2020-2021). https://www.epa.gov/system/files/documents/2022-12/2020-2021-biennial-
- 698 report.pdf (2022).
- 699 46. U.S. Environmental Protection Agency. Landfill Gas Emissions Model (LandGEM). (2023).
- 700 47. Feng, J., Li, Y., Strathmann, T. J. & Guest, J. S. Characterizing the Opportunity Space for
- Sustainable Hydrothermal Valorization of Wet Organic Wastes. *Environ. Sci. Technol.* **58**,
- 702 2528–2541 (2024).
- 48. Feng, J., Strathmann, T. & Guest, J. Financially Driven Hydrothermal-based Wastewater
- Solids Management for Targeted Resource Recovery and Decarbonization in the Contiguous
- 705 U.S. Preprint at https://doi.org/10.26434/chemrxiv-2025-qfxwd (2025).
- 706 707
- 708

Table of Contents

1. Supplementary Results	S2
2. Supplementary Methods	S9
2.1. General statement	S9
2.2. Treatment train assignments	S9
2.2.1. Description of treatment trains	S9
2.2.2. Assigning treatment trains to CWNS facilities	S12
2.2.3. Multiple treatment train assignments	S18
2.3. Treatment train energy requirements	S19
2.3.1. Energy requirement calculation	S19
2.3.2. Modified treatment train - *G1e	S20
2.3.3. New Treatment Trains	S20
2.3.4. Electricity carbon intensity	S36
2.4. Non-combustion on-site emissions of CH ₄ , N ₂ O, and CO ₂	S36
2.5. Biosolids handling	S37
2.5.1. Biosolids production and disposal	S37
2.5.2. Biosolids emission factors	S41
2.6. Uncertainty and sensitivity analyses	S43
2.6.1. Uncertainty values reported in main text	S45
2.7. Data access	S46
3. Supplementary Note 1: fossil origin carbon in wastewater	S47

1. Supplementary Results

1

2 3

4

5

6

7

8

9

1011

12

13

14

15

16

17

18

19

2021

22

23

Table S1. Summary of recent national-level wastewater treatment plan greenhouse gas inventories.

	European	China ²	China ³	U.S. ⁴	U.S.			
	Union ¹				(this work)			
Methodology								
	Bottom-up ^a	Bottom-up ^a	Bottom-up ^a	Top-down ^b	Bottom-up ^a			
Emissions breakdown ^c								
Electricity generation	Y (2)	Y (2)	Y (2)	N	Y (2)			
Natural gas	N	N	N	N	Y (2)			
Biological process CH ₄	Y (1)	Y (1, 2)	Y (2)	Y (1, 2)	Y (1, 2)			
Biological process N ₂ O	Y (1, 2)	Y (1, 2)	Y (2)	Y (1, 2)	Y (1, 2)			
Onsite non-combustion CO ₂ (biogenic)	N	Y ^d (2)	N	N	N			
Onsite non-combustion CO ₂ (fossil)	N	Y ^d (2)	N	N	Y (2)			
Sludge handling	Y (2)	N	N	Y (2)	Y (1,2)			
Sludge disposal	N	N	N	N	Y (1, 3)			
Effluent discharge	Y (1)	Y (1)	N	Y (1)	N			
Infrastructure	Y (2)	N	N	N	N			
Results								
Flow (billion m³/year)	N/A	67 ^e	65	N/A	67			
Emission (million tonne CO ₂ -eq/year)	34	30e	56	35	42			

^a Bottom-up: facility-level emission data aggregated to national level emission.

The datasets produced by this work are available in spreadsheet format, also available in our online GitHub repository in the folder <u>supplementary_databases</u> and deposited at this DOI: 10.5281/zenodo.15857431. Tabulated results files include:

- 1. Supplementary Dataset A: Wastewater treatment inventory of facilities
- 2. **Supplementary Dataset B**: Greenhouse gas emissions results for each treatment train Includes count, flow, energy requirement, per volume greenhouse gas emissions, and annual GHG emissions
- 3. **Supplementary Dataset C**: Greenhouse gas emissions inventory, including facility level results including identifying information and greenhouse gas emissions for each facility

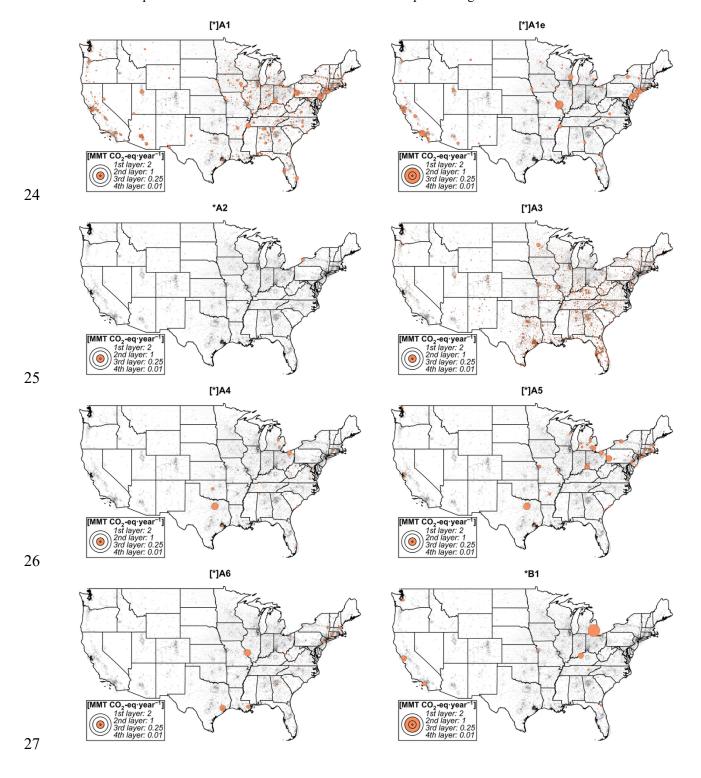
Uncertainty and Sensitivity Results: full results of our Monte Carlo simulation and Spearman's analysis are available in our online GitHub repository in the folder uncertainty sensitivity results.

^b Top-down: national level emission directly estimated based on national level flow rate or population data.

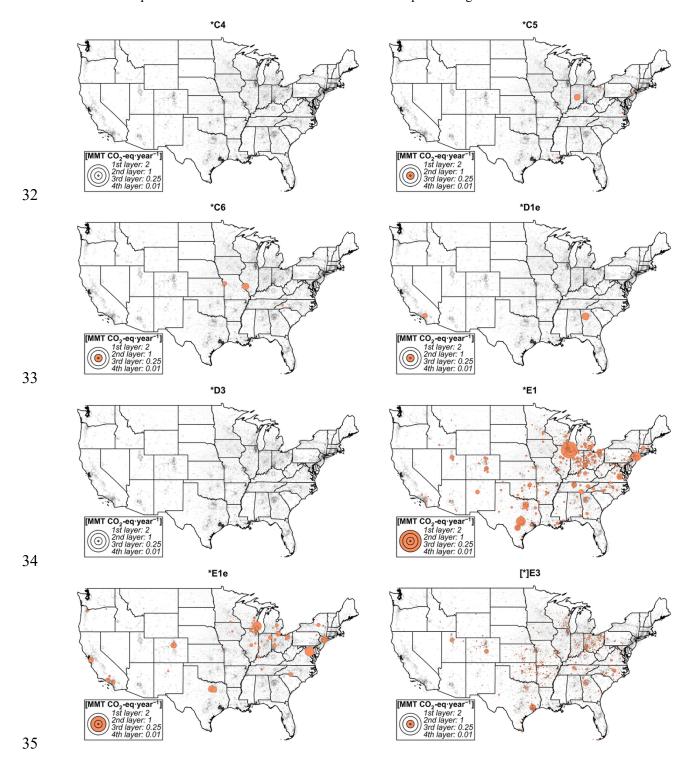
^c Y/N represents whether an emission type was included, followed by sources in the parenthesis (1= IPCC, 2=literature and/or government reports, 3=process modeling)

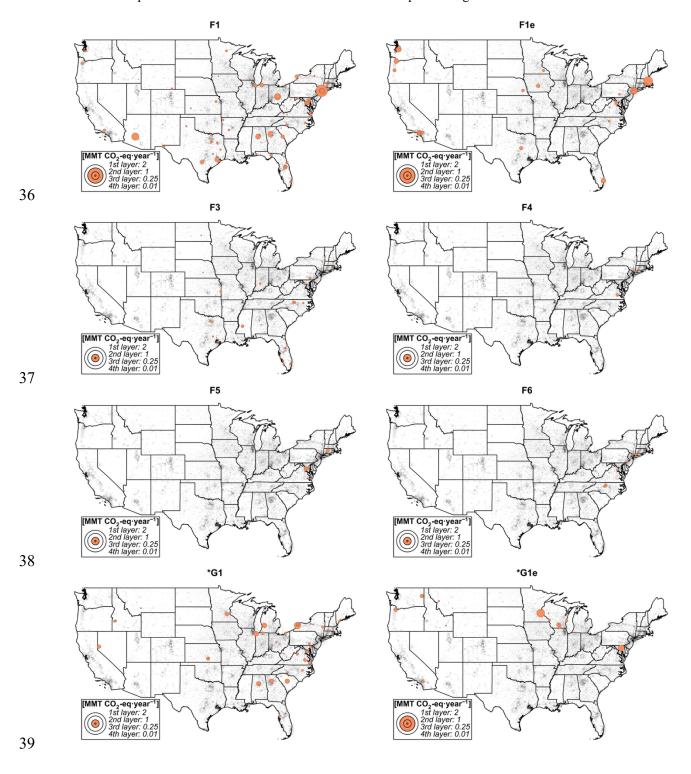
^d Fossil and biogenic emissions are not differentiated.

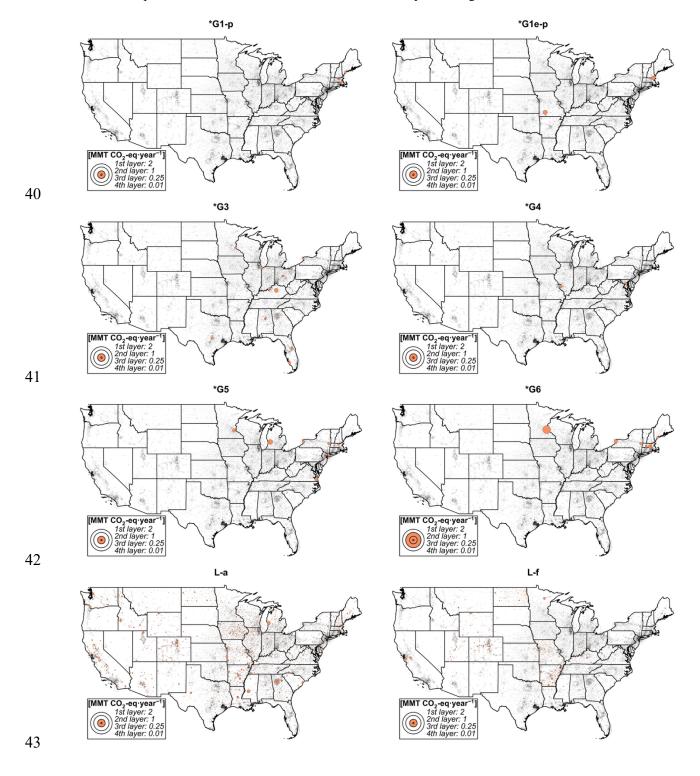
^e Flow and emission in 2019.











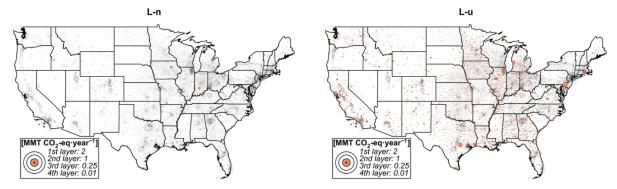


Figure S1. Geographic distribution of emissions from all treatment trains. Legends from inner to outer rings represent annual emission of 0.01, 0.25, 1, and 2 million metric tonne (MMT) CO₂-eq, respectively. Non-shaded legend indicates no wastewater treatment plant has emission in the corresponding level.

2. Supplementary Methods

2.1. General statement

All numbers reported in Sections 2.2 to 2.5 are baseline values without uncertainty. For detailed

uncertainty analysis, please refer to Section 2.6 and relevant methodology in the main manuscript.

2.2. Treatment train assignments

2.2.1. Description of treatment trains

We used Tarallo et al., 2015 as a baseline for establishing treatment trains for each facility in our database. However, to better represent the full range of wastewater treatment plant configurations in the United States, we supplemented the treatment trains defined in Tarallo et al., 2015 with additional combinations of liquids and solids processes. Table S2 and Table S3 provide a summary of key distinguishing features of the respective liquid and solids processes whose energy intensities were modeled using GPS-X in Tarallo et al., 2015. There are a total of seven different liquids treatment processes, which can occur with or without primary treatment and chemical phosphorous removal. There are also six different solids treatment configurations, one of which includes combined heat and power (CHP). Table S4 is a direct comparison of the treatment train codes employed in Tarallo et al., 2015 to the treatment train codes used in this work. Due to the frequent use of modeling results from Tarallo et al., 2015 in this document, we refer to all treatment trains using our treatment train naming convention followed by the Tarallo et al., 2015 code in parenthesis, sometimes abbreviated as a 'WERF' code.

Table S2. Liquid treatment codes with key parameters informing energy requirements. 'WERF Liquid Code' refers to the naming system used in Tarallo et al., 2015.

	WERF		Secondary Treatment			
Liquid Code (El Abbadi, Feng et al.)	Liquid Code (Tarallo et al., 2015)	Primary Treatment	Treatment Objective	Reactor Design	Chemical Phosphorous Removal	Chemical Inputs
A	С	No	Organics Removal	Activated Sludge – Basic	No	Hypochlorite
*A	В	Yes	Organics Removal	Activated Sludge – Basic	No	Hypochlorite
*B	0	Yes	Organics Removal	Activate Sludge – Pure Oxygen	No	Hypochlorite
*C	D	Yes	Organics Removal	Trickling Filter	No	Hypochlorite
*D	N	Yes	Phosphorous Removal	Membrane Bioreactor	No	Hypochlorite, Acetic Acid
Е	Е	No	Nitrification	Activated Sludge - Nitrification	No	Hypochlorite
*E	F	Yes	Nitrification	Activated Sludge - Nitrification	No	Hypochlorite

F	I	No	Nitrogen Removal	Activated Sludge - Biological Nutrient Removal (BNR)	No	Hypochlorite
*G	G	Yes	Phosphorous Removal	Activated Sludge - BNR	No	Hypochlorite, Acetic Acid
*G-p	Н	Yes	Phosphorous Removal	Activated Sludge - BNR	Yes	Hypochlorite, Acetic Acid

72 73

Table S3. Solids treatment codes with key parameters informing energy requirements. 'WERF Solids Code' refers to the naming system used in Tarallo et al., 2015.

Solids Code (El Abbadi, Feng et al.)	WERF Liquid Code (Tarallo et al., 2015)	Solids Stabilization	Recirculation	Chemical Inputs	Natural Gas Requirements	Biogas Use
1	1	Anaerobic digestion	After digestion and subsequent dewatering	None	Building heating	Building heat; anaerobic digester heating; excess gas is flared
1e	le	Anaerobic digestion + CHP	After digestion and subsequent dewatering	None	None†	Building heat; anaerobic digester heating; power generation from biogas offsets electricity requirements
2	4	Anaerobic digestion + direct thermal drying	After digestion and subsequent dewatering	None	Direct thermal drying	Building heat; anaerobic digester heating; direct thermal drying (alongside grid natural gas)
3	2	Aerobic digestion	After digestion and subsequent dewatering	None	None	N/A
4	3	Lime stabilization (Class B)	Dewatering and then recirculation before lime stabilization	Lime	Building heating; lime production	N/A
5	5	Multiple hearth incineration	Dewatering and then recirculation before incinera tion	None	Building heating; incineration	N/A

6	6	Fluidized bed incinerator	Dewatering and then recirculation before incineration	None	Building heating; incineration	N/A
---	---	---------------------------	---	------	--------------------------------------	-----

Note that for treatment train *G1e (G1E), we modified the WERF energy calculations to assume that all biogas is used to offset natural gas requirements from the grid.

Table S4. Comparison between treatment train codes used in this paper and those used by Tarallo et al., 2015.

El Abbadi, Feng et al. code	WERF code (Tarallo et al., 2015)
*A1	B1
*Ale	B1E
*A3	B2
*A4	B3
*A2	B4
*A5	B5
*A6	B6
Al	C1
Ale	C1E
A3	C2
A4	C3
A5	C5
A6	C6
*C1	D1
*Cle	D1E
*C3	D2
*C4	D3
*C5	D5
*C6	D6
E3	E2
*E3	E2P
*E1	F1
*E1e	F1E
*G1	G1
*Gle	G1E
*G3	G2
*G4	G3
*G5	G5
*G6	G6
*G1-p	H1
*Gle-p	H1E

77 78

F1	I1
Fle	I1E
F3	12
F4	13
F5	15
F6	16
*D1	N1
*D1e	N1E
*D3	N2
*B1	O1
*Ble	O1E
*B3	O2
*B4	O3
*B5	O5
*B6	O6
L-a	LAGOON_AER
L-n	LAGOON_ANAER
L-f	LAGOON_FAC
L-u	STBL_POND/LAGOON_OTHER

80 81

82

83 84

85

8687

88

2.2.2. Assigning treatment trains to CWNS facilities

Creating list of cumulative unit processes

We used data from the U.S. EPA's Clean Watersheds Needs Survey (CWNS)⁶⁻⁹ to identify active wastewater treatment plants in the United States, and the treatment processes present at these facilities as of 2022. Because not all facilities reported updated unit processes in the 2022 CWNS, we used the 2022 data in tandem with past surveys from 2004, 2008, and 2012 to form a more complete dataset of active treatment processes. When more granular or up-to-date external datasets were available, such as those regarding biogas utilization and treatment lagoons, we supplemented the unit processes reported in CWNS, as elaborated below.

899091

92

93

94

95

96

97

98

99

100

101

102

To develop a cumulative list of unit processes at each facility, we first aggregated unit process data across all CWNS surveys. We removed both duplicate unit processes and unit processes from 2008 and 2012 flagged for abandonment. In CWNS 2004, 2008, and 2012, flags indicating nutrient removal for each facility are reported separately from unit processes. We use these indicators (labeled in CWNS as: 'PRES AMMONIA REMOVAL,' 'PRES NIT REMOVAL,' and 'PRES PHOSPHOROUS REMOVAL') in addition to reported unit processes to identify facilities with nitrification, biological nutrient removal, or phosphorous removal. In our final treatment train dataset (Supplementary File A), we use a modified unit process code 'NIT FLAG' distinguish the facilities where nitrification added 'PRES AMMONIA REMOVAL' field; however, it is worth noting that some facilities may have nitrification reported in the form of both the ammonia removal flag ('NIT FLAG') and the unit process ('NIT').

CWNS does not specify whether biogas is used for heating to offset natural gas combustion, or as a biogenic fuel to produce electricity. Thus, we cross-referenced two additional databases to identify facilities that utilize digester biogas to produce electricity: the U.S. Department of Energy's (DOE) Combined Heat and Power Installation Database and the Water Environment Foundation's (WEF) Water Resource Recovery Facilities Biogas Database. ^{10,11} The DOE CHP database includes CHP installations across multiple industries through 2024, which we filtered only include to wastewater treatment facilities with biogas utilization technology installed before 2022 prior to incorporating it into treatment train assignments. The WEF database was originally developed based on a 2013 survey, and we accessed the website prior to a recent update which removed the option for users to download national data. Though the WEF website recommends that users cite the database as being last updated in 2024, we cannot confirm whether the data has been updated past 2013 without scraping the new web interface. Consequently, we used data previously downloaded from the retired interface in assigning treatment trains, which only included CHP installations through 2013.

CWNS also includes data on the presence of lagoons at treatment facilities, but does not require reporting the specifications of lagoon operation. Thus, lagoons can be reported as aerated, anaerobic, or facultative, but some are simply reported as "Lagoon, Other". We supplemented CWNS data with EPA's Lagoon Inventory Dataset (2022) to identify any additional lagoon-based facilities not reported in CWNS, and to determine lagoon type where possible. Because lagoons are more common in smaller, rural wastewater treatment facilities, we conducted an additional manual check on facilities with lagoons which reported flow rates greater than 10 MGD (n = 55). Using publicly available information, we verified whether or not a lagoon was present at these facilities. In this manner, we removed treatment lagoons from 14 facilities. Facilities which have been modified with manual checks are noted accordingly in the "UP_ID_NOTE" column of the facility inventory dataset (Supplementary File A).

If a unit process was reported in a less recent CWNS release (e.g., 2004, 2008, or 2012) but not in the 2022 CWNS, it was still considered an active component of the facility. The exception to this is facilities that reported multiple, conflicting secondary and/or solids treatment processes across the four survey years. For these facilities, we retained only the most recently reported secondary and/or solids treatment process(es) to minimize facilities with multiple treatment train assignments.

- Reliability of CWNS unit process data
- Table S5 summarizes unit process data reported in CWNS across survey years. In 2004, the survey required reporting all unit processes at a facility, while in 2008 and 2012, the survey only required reporting changes to existing unit processes through the use of change codes indicating if a facility
- was upgraded, abandoned, etc. In our inventory, 54.5% of facilities last reported unit process data

in 2004 but changes that occurred through 2012 are in theory captured through use of data from the 2008 and 2012 surveys. 74.5% of all facilities in our inventory have data from 2012 or earlier. We note that there is room for user interpretation of change codes used in 2008 and 2012. For example, a given unit process may have change codes for both abandonment and upgrading, and other researchers may differ from the approach we use, outlined above. In the 2022 survey, EPA removed change codes and thus we assume all unit processes reported as existing for a given facility are active. However, because reporting any unit process was optional in 2022, only 11.5% of our inventory reflects this most recent data, and the remaining 74.5% of our inventory rely on composite data from the 2012 CWNS and previous surveys. Our use of other databases, as described above, aims to better reflect changes that occurred within the last decade.

Table S5. Unit process data reporting in U.S. EPA's CWNS.

CWNS Survey Year	Unit process reporting requirement	Percent of facilities in survey year inventory reporting unit process data in survey year	Percent of facilities in 2022 inventory that last reported unit processes in survey year
2000	Report all unit processes	92.0%	0.1%
2004	Report all unit processes	92.3%	54.5%
2008	Report changes only	19.6%	0.8%
2012	Report changes only	27.5%	19.1%
2022	Fully optional	11.5%	11.5%

Assigning treatment trains

By searching for key combinations of treatment processes in the cumulative unit process list, we assigned one or more treatment trains for facilities with sufficient unit process data. The supplementary datafile "cwns_to_tt_codes.xlsx" in the treatment_train_assignment directory of this project's GitHub outlines the conversion from specific CWNS unit process codes to solids and liquids treatment processes within treatment trains. For facilities with partial unit process data available, we assigned treatment trains based on the most common treatment train(s) of the same plant size and EPA region with specific unit processes present. Lastly, for the remaining facilities with insufficient unit process data available, we assigned treatment trains based on the most common treatment train(s) of the same plant size and EPA region (Table S6). If the most common treatment train for a particular plant size and EPA region utilizes biogas for electricity, the non-electricity producing version of that treatment train was assigned for facilities with partial or insufficient unit process information, except for facilities that explicitly flagged as producing electricity in one or more of the supplemental biogas databases. Figure S2 summarizes how treatment trains were assigned based on unit process data availability, and Table S7 breaks this down further by flow rate and EPA region.

Table S6. Most common treatment train(s) by facility size and EPA region in 2022.

		. , , ,
2022 Flow Rate (MGD)	EPA Region	Most Common Treatment Train

	1	L-a (LAGOON AER)
	2	, = /
	3	A3 (C2)
		A3 (C2)
	4	L-u (STBL_POND)
Less than 2	5	L-u (STBL_POND)
	6	L-u (STBL_POND)
	7	L-u (STBL_POND)
	8	L-f (LAGOON_FAC)
	9	L-u (STBL_POND)
	10	L-u (STBL_POND)
	1	*A1 (B1)
	2	*A1 (B1)
	3	*C1 (D1)
	4	A3 (C2)
2 4	5	*C1 (D1)
2 - 4	6	A3 (C2)
	7	*C1 (D1)
	8	L-a (LAGOON_AER)
	9	L-a (LAGOON_AER)/L-u (STBL_POND)
	10	L-a (LAGOON_AER)
	1	*A1 (B1)
	2	*A1 (B1)
	3	*C1 (D1)
	4	*C1 (D1)
	5	*E1 (F1)
4 - 7	6	A3 (C2)
	7	*C1 (D1)
	8	*C1 (D1)
	9	L-u (STBL POND)
	10	*A1 (B1)
	1	*A1 (B1)
	2	*C1 (D1)
	3	*A1 (B1)
	4	*A3 (B2)
7 - 16	5	*E1 (F1)
	6	A3 (C2)
	7	*C1 (D1)
	8	*C1 (D1)
	9	*A1 (B1)
	10	*A1 (B1)
	1	*A5 (B5)/*A6 (B6)
16 - 46	2	*A1 (B1) *G1 (G1)/F1 (I1)

	4	*A1 (B1)
	5	*E1 (F1)
	6	*E1 (F1)
	7	*A1 (B1)
	8	*C1 (D1)
	9	*A1 (B1)
	10	*A1 (B1)/F1 (I1)
	1	*G6 (G6)/*G1-p (H1)/F5 (I5)
	2	*A1 (B1)
	3	F1 (I1)
	4	F1 (I1)
46 - 100	5	*E1 (F1)
	6	*E1 (F1)
	7	*C1 (D1)
	8	*C1 (D1)
	9	*A1 (B1)
	10	*G1 (G1)/F1 (I1)/*B1 (O1)
	1	F1 (I1)
	2	*E1 (F1)/F1 (I1)
	3	*A1 (B1)
	4	*B1 (O1)
C 4 1 100	5	*E1 (F1)
Greater than 100	6	*E1 (F1)
	7	*A6 (B6)/*C1 (D1)/*C6 (D6)
	8	*E1 (F1)
	9	*B1 (O1)
	10	F1 (I1)

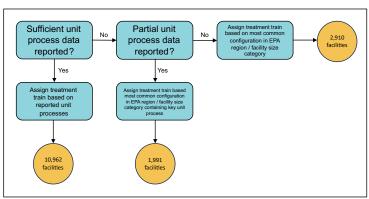


Figure S2. Breakdown of treatment train assignment based on unit process availability. Sufficient unit process data refers to one or more of the combinations of unit processes detailed in the treatment_train_werf() function of the tt_assignment_2022.ipynb script, available on this study's public GitHub repository. Partial unit process data refers to one or more of the following processes: basic activated sludge, activated sludge with nitrogen removal, activated sludge with phosphorous removal, activated

sludge with pure oxygen, anaerobic digestion, aerobic digestion, lime stabilization, fluidized bed incineration, multiple hearth incineration, trickling filters, or nitrification.

Table S7. Breakdown of treatment train assignment mechanism based on availability of unit process data, flow rate, and EPA region.

EPA Region	Sufficient Unit Process Information		Partial Unit Process Information		Insufficient Unit Process Information	
	Count	Total Flow (MGD)	Count	Total Flow (MGD)	Count	Total Flow (MGD)
1	356	1,734.639	124	324.885	69	77.262
2	480	4,688.991	116	633.427	183	96.478
3	616	3,576.490	116	234.958	179	127.540
4	1,305	6,626.355	309	921.813	442	1,020.949
5	2,871	9,448.227	382	591.619	391	503.128
6	1,603	4,534.983	558	823.167	623	957.418
7	2,017	2,712.010	208	169.266	511	38.721
8	796	1,229.560	50	172.788	272	152.987
9	424	3,782.210	76	703.427	145	966.682
10	494	1,691.528	52	116.430	95	28.654
Total	10,962	40,024.993	1,991	4,691.780	2,910	3,969.819

Because facilities often do not report every single unit process required for a particular treatment train configuration, treatment train assignments were made based on the presence or absence of a key subset of unit processes for each configuration. For instance, the treatment train *D3 (N2) is assigned when a facility reports both a membrane bioreactor and aerobic digestion. With this logic, some facilities may report enough unit processes to obtain multiple different treatment train assignments. For example, a facility in CWNS may report both pure oxygen activated sludge and basic activated sludge. We consider it likely that both processes were reported to describe the same pure oxygen activated sludge system, rather than the possibility that a facility contains two separate activated sludge systems, one using oxygen and one using air. Thus, we prioritized the assignment of treatment trains based on liquids process in the following order, an approach that also allows us to minimize the number of facilities assigned multiple treatment trains:

- 1. Activated sludge biological nutrient removal, phosphorus;
- 2. Activated sludge biological nutrient removal, nitrogen;
- 3. Nitrification;
- 4. Pure-oxygen activated sludge; and
- 5. Basic activated sludge.

Using the example described above, if a facility were to report all the unit processes required for both *B5 (O5) and *A5 (B5), it would only be assigned *B5 (O5), i.e. we assumed that a facility does not have both basic activated sludge and pure oxygen activated sludge, but rather all activated sludge facilities onsite use pure oxygen. The same logic applies for activated sludge-based nutrient removal systems. For additional information on treatment train assignment methodology, please see tt assignments 2022.ipynb posted on the public GitHub repository for this analysis.

2.2.3. Multiple treatment train assignments

Using the treatment train assignment methodology described above, it was possible for multiple treatment trains to be equally well-matched for a single facility, even after unit processes flagged for abandonment and outdated secondary/solids unit processes were removed from the cumulative unit process list. We found a total of 1,501 facilities that could be assigned multiple treatment trains. For instance, the Detroit Sewage Treatment Plant reports activated sludge, anaerobic digestion, chemical phosphorus removal, multiple hearth incineration, a configuration not accounted for in Tarallo et al., 2015. Subsequently, our assignment function assigned this facility treatment trains *B1 (O1) and *B5 (O5). Another notable instance of a facility receiving multiple treatment train assignments is that of nitrifying trickling filters, a configuration also not accounted for in Tarallo et al., 2015. Because our treatment train assignment methodology accounts for facilities that use a trickling filter or nitrification, but not both, if both nitrification and a trickling filter were reported as unit processes for a given facility, it was assigned both a nitrifying (liquids code E) and trickling filter treatment train (liquids code C).

Out of the 1,501 facilities with multiple treatment train assignments, 351 facilities reported multiple key secondary and/or solids processes in the most recent survey available for that facility. The remaining facilities consisted of combinations of treatment trains involving one or multiple types of lagoons, or a trickling filter coupled with another liquids treatment method. The majority (63% of the facilities) of the 351 facilities with multiple secondary/solids processes were instances of both aerobic and anaerobic digesters being present in a single facility. Approximately 23% of facilities with multiple secondary/solids processes reported digestion in addition to incineration.

We conducted further manual verification on a selection of large and small facilities with multiple secondary and/or solids treatment processes. While not a representative sample, these checks confirm that facilities may, in reality, have multiple secondary/solids processes in parallel or in series. However, it is also possible the second process was reported by mistake. Because verification of all facilities would require substantial manual effort, we chose to limit the number of multiple treatment train assignments by removing outdated secondary/solids processes. More specifically, if a facility reported different secondary/solids treatments across different survey years, we retained the processes reported most recently and disregarded the less recently reported ones. For the remaining facilities that still had enough unit process information to be assigned multiple treatment trains, we assumed that flow is split evenly across all identified trains. Note that

facilities with multiple, conflicting solids processes in this category make up approximately 9% of the national fleet by volume of treated wastewater, or 2% of total facilities.

246247248

249

250251

252

253

254

245

2.3. Treatment train energy requirements

2.3.1. Energy requirement calculation

We calculated the required electricity and natural gas inputs for each treatment train using the unit process energy requirements for the modeled treatment trains reported in Tarallo et al., 2015.

Table S8 lists these unit processes grouped by liquids treatment, solids treatment, and plant-wide loads at a modeled plant. Energy requirement per volume of treated wastewater varies for individual pieces of equipment across the treatment trains based on treatment objectives, recirculation configurations, and the presence/absence of CHP.

255256257

258

Table S8. Key energy consuming processes used in calculating electricity and natural gas requirements for treatment trains.

	Liquid Stream Processes	Solids Processes	Plant-wide	
Electricity	Equipment Influent pump station Screening & grit removal Primary clarifiers Biological reactor (blowers/pump as necessary based on design configuration) Final clarifies & recirculated activated	Equipment Gravity thickener Mechanical thickener Stabilization process (digester, incinerator, lime stabilization unit) Side stream pump	Odor control Site lighting	
	sludge pumping Disinfection	Dewatering Drying		
	Chemicals Hypochlorite production Acetic acid production	Chemicals Lime production		
Natural Gas	Equipment (via boiler) Anaerobic digester heating Chemicals Acetic acid production	Chemicals Lime production	Building heating (via boiler)	

259260

261

262

263

In **Table S9**, we list all the treatment trains used in this analysis, grouped into categories based on how we determined energy values per volume of treated wastewater. 'WERF Trains' refers to the treatment configurations with full energy requirements modeled and reported in Tarallo et al., 2015. For one treatment train, *G1e (G1E), we used the energy intensity values reported in Tarallo

et al., 2015 with minor modifications. Finally, 'New Trains' refers to configurations that were not modeled in Tarallo et al., 2015 for which we used combinations of energy values for liquids and solids processes reported in Tarallo et al., 2015 with modifications documented below.

Table S9. Overview of all treatment trains used in our analysis, grouped based on whether they are WERF Trains reported in Tarallo et al., 2015, modified WERF trains, or new trains reported for this first time in this analysis.

	WERF Trains	Modified WERF Trains	New Trains
Feng et al.	*A1, *A1e, *A2, *A5, *A6, A4, *C1, E3, *E3, *E1, *G1, *G1-p, F4, *D1, *D3, *B1	*G1e	*A3, *A4, A1, A1e, A3, A5, A6, *C1e, *C3, *C4, *C5, *C6, *E1e, *G3, *G5, *G6, *G1e-p, F1, F1e, F5, F6, *D1e, *B1e, *B3, *B4, *B5, *B6
WERF Code	B1, B1E, B4, B5, B6, C3, D1, E2, E2P, F1, G1, H1, I2, I3, N1, N2, O1	G1E	B2, B3, C1, C1E, C2, C5, C6, D1E, D2, D3, D5, D6, G2, G3, F1E, G5, G6, H1E, I1, I1E, I5, I6, N1E, O1E, O2, O3, O5, O6

2.3.2. Modified treatment train - *G1e

We made slight modifications to the treatment train *G1e (G1E) energy requirements compared to values reported by Tarallo et al., 2015. In the reported configuration, *G1e includes power generation through CHP, as well as a natural gas input from the grid of 1,600 MJ/day. However, we noted that *G1e (G1E) is the only train reported in Tarallo et al., 2015 with CHP that also uses natural gas from the grid, all other CHP trains rely on biogas instead. The *G1 (G1) train, upon which *G1e (G1E) is based, does not import any natural gas use either. Thus, for our analysis we removed the imported natural gas for *G1e (G1E), and scaled back onsite electricity production accordingly. Specifically, the 1,600 MJ/day of natural gas from the grid was replaced by 1,600 MJ/day of biogas from the anaerobic digester diverted to the boiler instead of directed to the generator, as originally reported in Tarallo et al., 2015:

Reported Generator Efficiency =
$$\frac{5,760 \text{ kWh/day produced from generator}}{62,300 \text{ MJ/day into generator}} = 9.24\%$$

To account for the reduced electricity supply from the generator, we increased the electricity requirement from the grid accordingly.

2.3.3. New Treatment Trains

We calculated the energy requirements for new treatment trains using combinations of liquids and solids treatment processes reported by Tarallo et al., 2015, with modifications to specific unit

processes, as deemed necessary. **Table S10** summarizes our approach for deriving energy values from WERF treatment trains, a detailed discussion of which is provided below.

297 298 299

300

301

295

Table S10. Descriptions of how energy values were derived for new treatment trains in this study. We use our nomenclature with the corresponding WERF code included parenthetically. The process models from Tarallo et al., 2015 used as a baseline for determining energy requirements for new treatment train configurations are listed in the source configuration columns.

New Treatment Train	Liquid Process Source Configuration	Solids Stabilization Source Configuration
*A3 (B2)	*A1 (B1)/*A2 (B4) (identical liquids treatment energy requirements)	Aerobic digester energy calculated using influent chemical oxygen demand from *A1 (B1) as input to linear regression. Average thickener values for gravity + mechanical thickeners present.
*A4 (B3)	Average *A5 (B5) and *A6 (B6)	Lime production energy calculated using average dosing rate. Lime stabilization process energy using average of A4 (C3) and F4 (I3). Average thickener values for gravity + mechanical thickeners present.
A1 (C1)	A4 (C3)	Basic activated sludge cluster for anaerobic digestion (*A1 (B1)/*A1e (B1E)/*E1, P1, P1E). Average thickener values for mechanical thickener only present.
Ale (C1E)	A4 (C3)	Basic activated sludge cluster for anaerobic digestion (*A1 (B1)/*A1e (B1E)/*E1, P1, P1E). Average thickener values for mechanical thickener only present. Biogas produced based on linear regression of influent chemical oxygen demand. Electricity generated based on biogas produced and generator efficiency of 0.0925.
A3 (C2)	A4 (C3)	Aerobic digester energy calculated using influent chemical oxygen demand from A4 (C3) as input to linear regression. Thickener average values for mechanical thickener only present.
A5 (C5)	A4 (C3)	*A5 (B5) for incinerator. Thickener average values for mechanical thickener only present.
A6 (C6)	A4 (C3)	*A6 (B6) for incinerator. Thickener average values for mechanical thickener only present.
*Cle (D1E)	*C1 (D1)	Electricity generated based on biogas produced and generator efficiency of 0.0925.
*C3 (D2)	*C1 (D1)	Aerobic digester energy calculated using influent chemical oxygen

		demand from *C1 (D1) as input to linear regression. Thickener average values for mechanical thickener only present.
*C4 (D3)	*C1 (D1)	Lime production energy calculated using average dosing rate. Lime stabilization process energy using average of A4 (C3) and F4 (I3). Average thickener values for gravity thickener only.
*C5 (D5)	*C1 (D1)	*A5 (B5) for incinerator. Thickener average values for gravity thickener only present.
*C6 (D6)	*C1 (D1)	*A6 (B6) for incinerator. Thickener average values for gravity thickener only present.
*E1e (F1E)	*E1 (F1)	Electricity generated based on biogas produced and generator efficiency of 0.0925.
*G3 (G2)	*G1 (G1)/*G1e (G1E) (identical liquids treatment energy requirements)	Aerobic digester energy calculated using influent chemical oxygen demand from *G1 (G1) as input to linear regression. Average thickener values for gravity + mechanical thickeners present.
*G4 (G3)	*G1 (G1)/*G1e (G1E) (identical liquids treatment energy requirements)	Lime production energy calculated using average dosing rate. Lime stabilization process energy using average of A4 (C3) and F4 (I3). Average thickener values for gravity + mechanical thickeners present.
*G5 (G5)	*G1 (G1)/*G1e (G1E) (identical liquids treatment energy requirements)	*A5 (B5) for incinerator. Average thickener values for gravity + mechanical thickeners present.
*G6 (G6)	*G1 (G1)/*G1e (G1E) (identical liquids treatment energy requirements)	*A6 (B6) for incinerator. Average thickener values for gravity + mechanical thickeners present.
*G1e-p (H1E)	*G1-p (H1)	Electricity generated based on biogas produced and generator efficiency of 0.0925.
F1 (I1)	F3 (I2)	Nutrient Removal anaerobic digestion cluster (*G1 (G1)/*G1e (G1E)/*G1-p (H1)/*D1 (N1)) for anaerobic digester energy. Thickener average values for mechanical thickener only present.
Fle (I1E)	F3 (I2)	Nutrient Removal anaerobic digestion cluster (*G1 (G1)/*G1e (G1E)/*G1-p (H1)/*D1 (N1)) for anaerobic digester energy. Thickener average values for mechanical thickener only present.

		CHP energy production based on *G1e (G1E).
F5 (I5)	F4 (I3)	*A5 (B5) for incinerator. Thickener average values for mechanical thickener only present.
F6 (I6)	F4 (I3)	*A6 (B6) for incinerator. Thickener average values for mechanical thickener only present.
*D1e (N1E)	*D1 (N1)	Electricity generated based on biogas produced and generator efficiency of 0.0925.
*B1e (O1E)	*B1 (O1)	*B1 (O1). Energy production from *A1e (B1E).
*B3 (O2)	*B1 (O1)	Aerobic digester energy calculated using influent chemical oxygen demand from *B1 (O1) as input to linear regression. Thickener average values for gravity thickener only present.
*B4 (O3)	*B1 (O1)	Lime production energy calculated using average dosing rate. Lime stabilization process energy using average of A4 (C3) and F4 (I3). Thickener average values for gravity thickener only present.
*B5 (O5)	*B1 (O1)	*A5 (B5) for incinerator. Thickener average values for gravity thickener only present.
*B6 (O6)	*B1 (O1)	*A6 (B6) for incinerator. Thickener average values for gravity thickener only present

Liquids treatment processes

302303

304

305

306

307308

309

To determine the energy requirement of the liquids process of a new treatment train, we used the energy requirements from a 'base train', a WERF train with reported energy values in Tarallo et al., 2015. A base train will have the same liquid treatment process as the corresponding new train, but a different solids treatment process. **Figure S3** summarizes the logic for selecting the base train for the liquid treatment process.

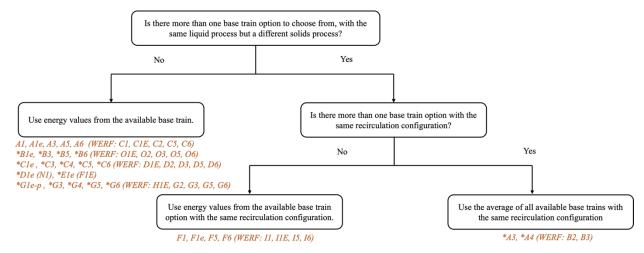


Figure S3. Logic for selecting the base train for liquids treatment train processes in all new trains. Relevant trains for each approach are listed in orange. We identified all WERF trains with the same liquid treatment process, and, when possible, selected a base train based on similar side-stream recirculation configurations.

When only one base train option was available to choose from, we used the reported values for the corresponding new train's liquid process. For example, A4 (C3) is the only train with this particular liquid configuration with reported energy values. Thus, when calculating energy requirements for the liquid portion of A1 (C1), A1e (C1E), A3 (C2), A6 (C5) and A6 (C6), we must take the A train values reported in A4 (C3). We apply this approach for the following trains: A1 (C1), A1e (C1E), A3 (C2), A6 (C5), A6 (C6), *C1e (D1E), *C3 (D2), *C4 (D3), *C5 (D5), *C6 (D6), *E1e (F1E),*G3 (G2), *G4 (G3), *G5 (G5), *G6 (G6), *G1e-p (H1E), *D1e (N1E), *B1e (O1E), *B3 (O2), *B4 (O3), *B5 (O5), and *B6 (O6).

However, the energy requirements within the liquids treatment portion of a train may vary with downstream solids treatment processes. Thus, where more than one base train option was available, we selected the base train for the new train configuration based on side-stream recirculation associated with downstream solids processing. Side-stream composition and solids content is impacted by whether or not recirculation occurs before or after the solids treatment process. In the models used by Tarallo et al., 2015, dewatering and recirculation occur *before* incineration and lime stabilization, but for aerobic and anaerobic digestion, the solids stream is dewatered *after* the stabilization in the digester (see **Figure S4**).

Solids Stabilization Configurations	<u>Code</u>
Anaerobic Digestion ——— Dewatering	1
Anaerobic Digestion ——— Dewatering ——— Direct Thermal Drying	2
Aaerobic Digestion ——— Dewatering	3
Dewatering — Lime Stabilization	4
Dewatering ——→ Multiple Hearth Incineratio	n 5
Dewatering ——→Fluidized Bed Incineration	6

Figure S4. Solids stabilization configurations with dewatering sequence.

For example, when determining the energy requirements for F1 (I1), there are two liquid treatment train options to choose from: the F train coupled with aerobic digestion, F3 (I2), or lime stabilization, F4 (I3). We choose to draw values from the configuration with aerobic digestion, rather than lime stabilization, for closer alignment in recirculation configuration which can affect the energy requirements of the unit processes downstream of recirculation (biological reactor blowers, final clarifiers, recirculating activated sludge pumping). Finally, if multiple treatment trains are available to select from with the same recirculation configuration, we used the average value of the energy requirements from these trains. Note that this only occurred when calculating energy requirements for *A3 (B2) and *A4 (B3).

Solids treatment processes

Energy requirements for solids treatment processes vary based on the upstream treatment. Consequently, we used data reported in Tarallo et al., 2015 to extrapolate electricity and natural gas requirements for the solids treatment component of the new treatment trains.

Aerobic digestion. Using data reported by Tarallo et al., 2015, we found a linear relationship between aerobic digester requirements and influent chemical oxygen demand (COD), the embedded chemical energy of wastewater. This is consistent with the typical assumption that electricity consumption is driven by aeration to support COD degradation. For each new treatment train, we calculated the expected influent COD to the aerobic digester using the COD flow rate (reported by Tarallo et al., 2015 in MJ/day) leaving the mechanical and/or gravity thickeners, as reported for the liquid base train. For example, we used COD leaving the gravity and mechanical thickeners in *A1 (B1) to determine the influent COD to the aerobic digester in *A3 (B2). Using the linear best-fit for the WERF trains (Figure S5), we calculated electricity consumption for each new train (Table S11).

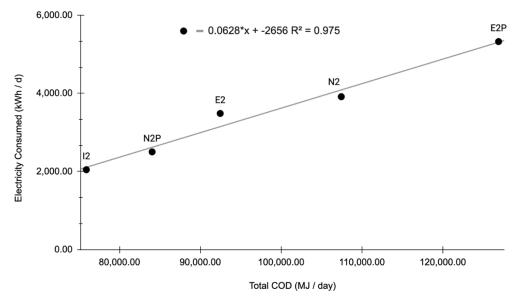


Figure S5. Electricity consumption for aerobic digestion as a function of total COD into the digester. Note that the x-axis starts at 75,000 MJ/day.

Table S11. Calculated electricity consumption for new treatment trains with aerobic digestion for solids treatment.

New Train	Liquid Train	Solids Train	COD to Digester (MJ/d)	Electricity Consumed by Digester (kWh/d)
*A3 (B2)	*A1 (B1)	*E3 (E2P)	126,439.00	5,284.37
A3 (C2)	A4 (C3)	*D3 (N2)	109,939.00	4,248.17
*C3 (D2)	*C1 (D1)	*D3 (N2)	108,205.00	4,139.27
*G3 (G2)	*G1 (G1)	*E3 (E2P)	128,460.00	5,411.29
*B3 (O2)	*B1 (O1)	*E3 (E2P)	128,653.00	5,423.41

Anaerobic digestion: electricity requirement

We did not observe a linear relationship between electricity required for anaerobic digestion in WERF modeled treatment trains and available parameters, such as influent COD (**Figure S6**) or bioreactor volume (**Figure S7**). However, electricity requirements appear to cluster roughly into groups, with lower electricity for trains without nutrient removal (*A1 (B1), *C1 (D1), *B1 (O1), and WERF trains A1 and P1 which are not included in our analysis) and greater electricity needed for nutrient removal (*G1-p (H1), *G1 (G1), *D1 (N1)). Thus, to create treatment trains A1/A1e (C1/C1E) (activated sludge without primary treatment), we used the electricity requirement from *A1 (B1), activated sludge with primary treatment. For F1 (I1), biological nitrogen removal activated sludge without primary treatment, we used the *G1-p (H1)/*G1 (G1)/*D1 (N1) energy requirement, all of which are equivalent.

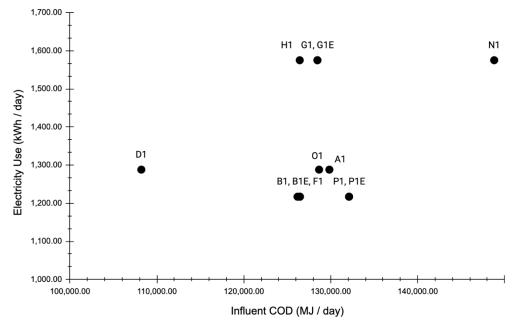


Figure S6. Electricity consumption as a function of influent COD for anaerobic digestion treatment trains reported in Tarallo et al., 2015. Note that the x and y-axes start at 100,000 MJ COD / day and 1,000 kWh/day respectively.

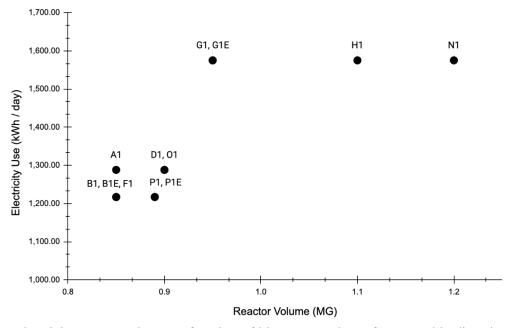


Figure S7. Electricity consumption as a function of bioreactor volume for anaerobic digestion treatment trains reported in Tarallo et al., 2015. Note that the y-axis starts at 1,000 kWh/day.

 Anaerobic digestion: biogas production. In the WERF treatment trains, we observed a linear relationship between biogas production and influent COD. We used a linear regression with a fixed y-intercept at the origin to determine the average biogas production per unit of COD influent, as depicted in Figure S8. From these regression results, we calculated biogas production for new treatment trains A1e (C1E) and F1e (I1E) using the estimated influent COD to the anaerobic digester, a value dependent on the liquid train the new treatment train is based on. For A1e (C1E), we approximated the influent COD to the anaerobic digester as the effluent COD from the mechanical thickener of A4 (C3). For F1e (I1E), the influent COD is approximated as the effluent COD of the mechanical thickener of F3 (I2). For new treatment trains *C1e (D1E), *E1e (F1E), *G1e-p (H1E), *D1e (N1E) and *B1e (O1E), we used the biogas production reported for the non-electricity producing version of the treatment train published in Tarallo et al., 2015. Influent COD and predicted biogas production are included in Table S12.

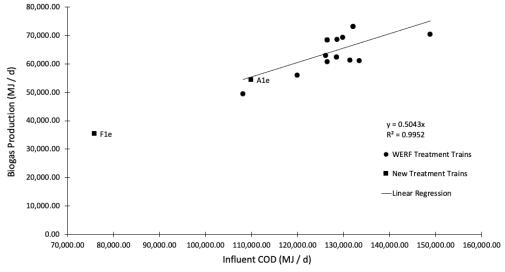


Figure S8. Linear regression used to estimate daily biogas production. For new treatment trains A1e (C1E) and F1e (I1E), we calculated biogas production based on the influent COD to the digester. For treatment trains *C1e (D1E), *E1e (F1E), *G1e-p (H1E), *B1e (O1E), and *D1e (N1E), we used the biogas production rate reported in Tarallo et al., 2015 for the non-electricity producing versions of these trains. Note that the x-axis starts at 70,000 MJ / day.

Table S12. Predicted daily biogas yield for new treatment trains.

	Influent COD (MJ/day)	Predicted Biogas Production (MJ/day)
Ale (C1E)	109,939	58,484
*C1e (D1E)	108,205	49,300
*E1e (F1E)	126,173	62,900
*G1e-p (H1E)	126,428	60,700
F1e (I1E)	75,899	45,986
*D1e (N1E)	148,796	70,400

*B1e (O1E)	128,653	68,500

Anaerobic digestion: energy generation. Our analysis includes seven new treatment trains with CHP for onsite electricity production: A1e (C1E), *C1E (D1E), *E1e (F1E), *G1e-p (H1E), F1e (I1E), *D1e (N1E), and *B1e (O1E). WERF provides modeled energy values for three treatment trains with CHP: *A1e (B1E), *G1e (G1E) and P1E, all of which produce different amounts of energy per day. Thus, we assigned energy production values for new treatment trains based on the estimated amount of biogas produced and an average generator efficiency of 0.0925, calculated based on the WERF treatment trains that utilize biogas for electricity generation.

Lime stabilization. We determined energy requirements for lime stabilization using the assumptions reported by Tarallo et al., 2015. Following their approach, we assume Class B lime stabilization is used to achieve a reduced pathogen level, as opposed to complete removal. Energy requirements for lime stabilization treatment trains can be broken up into two components: the natural gas and electricity needed to produce the lime itself (typically offsite), and electricity needed onsite to operate the lime stabilization unit. For the process electricity required to operate the lime stabilization unit, we use the average of the two reported WERF trains with lime stabilization, A4 (C3) and F4 (I3).

Tarallo et al., 2015 report the electricity and natural gas required to produce lime offsite (**Table S13**) and assume a dose of 85 lbs of lime per ton of wet weight into the lime stabilization unit. However, mass flow data is only reported in total suspended solids (TSS) concentration (lbs TSS/day) and flow rate (MGD/day) entering the lime stabilization unit, and wet weight is not provided. Subsequently, to estimate the energy needed for lime production for our new treatment trains *A4 (B3), *C4 (D3), *G4 (G3), and *B4 (O3), we calculated lime dosing as a function of TSS using provided energy values. Because Tarallo et al., 2015 process model documentation (provided by the authors for use in this work) only includes supplemental mass balance data with TSS concentration for F4 (I3) and not A4 (C3), we use the energy requirements for lime stabilization reported in F4 (I3) to calculate the amount of lime used per day, and in turn the amount of lime needed per lb of TSS. Calculations are included in **Table S14**.

Table S13. Energy requirements for lime production reported in Tarallo et al., 2015 and used as model inputs.

Energy input for lime production	Quantity
Electricity (kWh/lb lime)	0.028056
Natural gas (MJ/lb lime)	2.3319

Table S14. Calculations for determining lime dosage as a function of TSS.

F4 (I3) Lime Calculatio	ns	
Reported Fuel Used for Chemical Production	MJ/day	7,178.48

Reported Electricity Used for Chemical Production	kWh/day	86.3657
Calculated lime production/day - using electricity	lbs lime/day	3,078
Calculated lime production/day - using fuel	lbs lime/day	3,078
Reported cake production	lbs TSS/day	12,314
Calculated lime dose	lbs lime/lb TSS	0.25

Because we only have full information for calculating lime dosing and energy requirements for one treatment train, we examined the potential variability if lime stabilization were used across all treatment trains. Thus, using available cake concentrations and flow rates reported by Tarallo et al., 2015, we estimated the total amount of lime needed for stabilization for the treatment trains showed in **Table S15** and calculated the corresponding natural gas and electricity requirements using the dosing rate calculated in **Table S14**. **Table S15** includes values for all treatment trains, and **Table S16** reports summary statistics. We used the average energy values for lime production from across all modeled WERF trains for the new treatment trains with lime stabilization.

Table S15. Predicted electricity and natural gas required to stabilize the cake generated from the WERF treatment train using lime. We used the mass balance data available for WERF trains to estimate the energy requirements based on the mass of the cake leaving the dewatering process across all configurations. For the treatment train naming system, we list our code first, followed by the Tarallo et al., 2015 code in parenthesis. Treatment trains with only values in parenthesis are those which are not included in our analysis.

Treatment Train	Electricity Required (kWh/d)	Natural Gas Required (MJ/d)
(A1)	67.21	5,586.49
*A5 (B5)	126.22	10,490.66
*A6 (B6)	126.14	10,483.85
*A1 (B1)	64.53	5,363.17
*A1e (B1E)	64.53	5,363.17
*A2 (B4)	64.53	5,363.17
E3 (E2)	69.46	5,773.22
*E3 (E2P)	81.52	6,775.48
*E1	69.68	5,791.42
*G1 (G1)	79.90	6,640.90
*G1e (G1E)	79.89	6,640.43
*G1-p (H1)	95.75	7,958.76
F3 (I2)	67.16	5,582.16
F4 (I3)	86.37	7,178.48
(L1)	117.29	9,748.99

(M1)	114.27	9,497.64
*D1 (N1)	95.89	7,970.31
*D3 (N2)	88.43	7,349.69
A4 (C3)	109.00	9,030.00
Average	87.78	7,294.10

Table S16. Summary statistics for energy required for lime stabilization across all WERF trains. We used the mean energy requirement in all new trains with lime stabilization.

Expected Energy Requirement						
Electricity (kWh/day) Natural Gas (MJ/day						
Mean	87.78	7,294.08				
Minimum	64.53	5,363.17				
Maximum	126.22	10,490.66				
Standard Deviation	21.60	1,793.97				

 Incineration. Tarallo et al., 2015 only model two treatment trains with incineration, one with multiple hearth incineration (MHI) and one with fluidized bed incineration (FBI). Thus, for all new treatment trains with incineration, we use the electricity and natural gas required for incineration in *A5 (B5) (MHI) or *A6 (B6) (FBI), based on the respective type of incinerator in the new train.

Ancillary solids treatment processes. In addition to the main solids stabilization process, energy is required for ancillary processes such as solids thickening and dewatering. Across all reported solids treatment trains, there are minor fluctuations in energy requirements for gravity and mechanical thickener requirements. Therefore, for all new treatment trains, we use the average energy requirements for thickeners based on the following configurations: gravity thickener only, mechanical thickener only, and both thickeners present. Additionally, energy is used for dewatering, which remains constant across all WERF trains at 89 kWh/day. These values are summarized in **Table S17**.

Table S17. Energy requirements calculated for thickeners, inclusive of dewatering energy use.

Configuration	Gravity Thickener Energy (kWh/day)	Mechanical Thickener Energy (kWh/day)
Gravity Thickener Only	27.5	0.0
Mechanical Thickener Only	0.0	162.3
Both Thickeners Present	24.1	157.1

Energy calculations sensitivity analysis

We evaluated the sensitivity of total energy requirements to variation in the assumptions we made regarding energy requirements for individual components of the treatment. Specifically, we developed upper and lower sensitivity bounds for aerobic digestion, anaerobic digestion, and lime stabilization. However, we did not include any liquids processes or minor steps in the solids treatment processes (e.g. thickening), which exhibit minimal variation in energy inputs across the WERF treatment train model results. For all energy values calculated using linear regression (electricity consumption by aerobic digesters and electricity production from anaerobic digester biogas), we determined the associated 95% confidence intervals (CI) to use as upper and lower bounds on these inputs. For unit processes where energy values were selected based on similar treatment objectives (anaerobic digester electricity consumption), as well as lime stabilization, we used the lowest and highest energy requirements reported across all WERF treatment trains as the lower and upper bounds, respectively.

Aerobic digestion. To determine the uncertainty associated with energy required for aerobic digestion, we calculated 95% CIs on the linear regression used to determine the relationship between influent COD and electricity requirement (**Figure S9**). Upper and lower bounds of the 95% CI were used as the upper and lower bounds in our sensitivity analysis for the following relevant treatment trains: *A3 (B2), A3 (C2), *C3 (D2), *G3 (G2), and *B3 (O2).

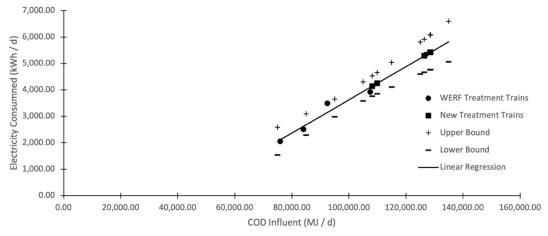


Figure S9. Uncertainty bounds of linear regression used to estimate electricity demand of aerobic digesters in sensitivity analysis. We calculated the 95% CI on electricity consumption values calculated using linear regression.

Anaerobic digestion: electricity. For upper and lower bounds on electricity demands for anaerobic digesters, we used the highest and lowest reported electricity value for trains modeled in Tarallo et al., 2015: *A1 (B1) and *G1 (G1), respectively. These bounds contribute to the sensitivity analysis for the following trains: A1 (C1), A1e (C1E), F1 (I1), F1e (I1E), and *B1e (O1E).

Anaerobic digestion: biogas production. To assess uncertainty in biogas production, we used the 95% CIs on the linear regression used to determine biogas production based on influent COD, applicable to treatment trains A1e (C1E) and F1e (I1E) (Figure S10). Upper and lower CIs are used as inputs for the sensitivity analysis. For the remaining treatment trains, modeled biogas production is reported in Tarallo et al., 2015 and thus we do not apply a sensitivity analysis.

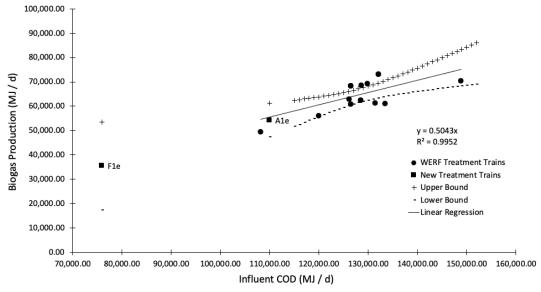


Figure S10. Uncertainty bounds of linear regression used to estimate biogas production of anaerobic digesters in sensitivity analysis.

Lime production. For lower and upper bounds on energy required for lime production, we used the treatment trains with the lowest and highest theoretical energy requirements for lime production, *A1 (B1) and *A5 (B5). For the electricity required for the process of lime stabilization itself, we used the energy requirements from the least and most energy intensive lime treatment trains, F4 (I3) and A4 (C3), as lower and upper bounds.

Sensitivity analysis results. Figure S11 depicts the ranges we observe in energy estimates across new treatment trains. With current assumptions, new trains involving lime production/stabilization or aerobic digesters have the greatest range of possible energy intensities. However, when compared to the range in energy intensities across all treatment trains, the variability associated with our assumptions is far less than the variability between different treatment trains.

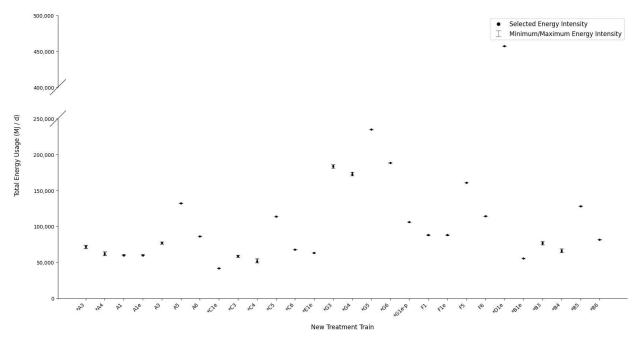


Figure S11. Sensitivity of total energy requirement to variability of model results from Tarallo et al., 2015. Lower and upper bounds were determined by considering the modeled range of energy requirements in each process of a treatment train. Where only the selected energy intensity point is visible, differences in values are too small to be apparent at a scale which allows for comparison between all treatment trains.

Figure S12 depicts a sensitivity analysis for the imported electricity requirement, which captures variability in our assumptions regarding energy generated onsite. This figure also captures the variability observed in energy requirements for aerobic digestions, more visible here when separated out from the natural gas requirements, which shown in isolation in Figure S13. We observed the greatest variability in treatment train F1e (I1E), reflective of the wide 95% CI associated with the linear regression used to determine biogas production. This high uncertainty reflects the fact that the influent COD for the F1E (I1E) treatment train is far lower than values reported for other WERF treatment trains with anaerobic digestion. However, even in this instance, the variability remains acceptable when considering the differences in imported energy requirements that exist between different treatment trains.

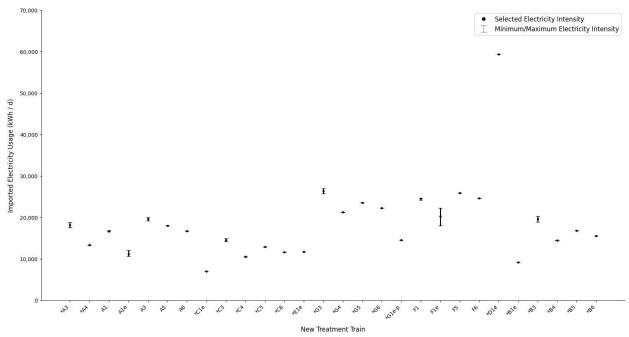


Figure S12. Sensitivity of imported electricity. Lower and upper bounds were determined by considering the modeled range of energy requirements in each process of a treatment train. For trains with anaerobic digestion, we also considered the range in biogas production rate and corresponding electricity consumption. Where only the selected energy intensity point is visible, differences in values are too small to be apparent at a scale which allows for comparison between all treatment trains.

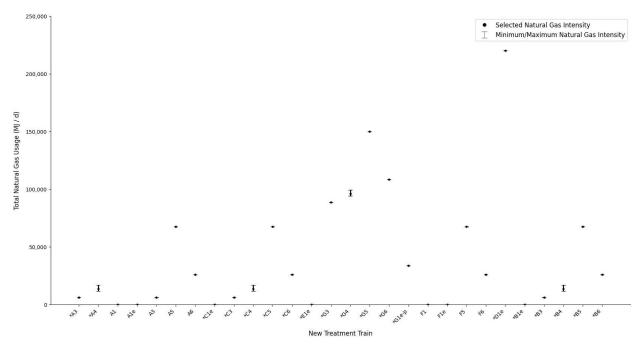


Figure S13. Sensitivity of imported natural gas. Lower and upper bounds were determined by considering the modeled range of energy requirements in each process of a treatment train. Where only the selected

energy intensity point is visible, differences in values are too small to be apparent at a scale which allows for comparison between all treatment trains.

563564565

566567

568

571

562

2.3.4. Electricity carbon intensity

Greenhouse gas (GHG) emission factors for production of different types of electricity were assigned according to values in **Table S18**. This table also includes emissions associated with producing and burning natural gas.

569570

Table S18. Greenhouse gas emissions factors for electricity by power plant type, and for natural gas, including emissions from extraction and distribution systems, power plant construction, and combustion. ¹⁵

Power plant type	GHG emissions (g CO2-eq/MJ)
Natural Gas	24
Coal	18
Nuclear	1.9
Wind	2.9
Solar	10
Biomass	19
Geothermal	1.4
Hydro	2.1
Natural gas system	GHG emissions (g CO ₂ -eq/MJ)
Extraction and distribution emissions	13
Combustion emissions	56

572573

574

575

2.4. Non-combustion on-site emissions of CH₄, N₂O, and CO₂

We developed CH₄ and N₂O factors emissions using values reported in literature. We converted all emissions estimates to equivalent CO₂ emissions based on their 100-year Global Warming Potential, 29.8 for methane and 273 for nitrous oxide.¹⁶

576577578

579

580

581

For CH₄ emissions (**Table S19**), we used emission factors from Song et al.¹⁷ based on the presence or absence of an anaerobic digester at the facility. For lagoons, we used IPCC emission factors for anaerobic, aerobic, and facultative lagoons. For the uncategorized lagoons emission factor, we used the national flow-weighted average of emission factors for aerobic, anaerobic, and facultative lagoons.¹⁸

Table S19. Baseline emission factors for methane.

Category	CH ₄ [g CH ₄ /m ³]
Facility with anaerobic digestion ¹⁷	11.4
Facility without anaerobic digestion ¹⁷	1.37
Aerobic lagoon ¹⁹	0
Anaerobic lagoon ¹⁹	25.4
Facultative lagoon ¹⁹	25.4
Uncategorized lagoon	4.56

To estimate nitrous oxide emissions (**Table S20**), we used the 376 nitrous oxide emission factor observations reported by Song et al., 2024.²⁰ Using their reported literature database, we categorized each measurement into a nutrient removal category (organics removal, nitrification, denitrification) based on their reported treatment configuration. Where the listed reactor design could be configured for multiple treatment objectives, we used the process descriptions provided in the original publications cited by Song et al. Next, we calculated emission factors for each treatment objective using measurements identified by Song et al. as being conducted at either bioreactor or facility level scale, and from full-scale facilities only (i.e. excluding emission factors from pilot scale facilities). Of the 281 reported emissions factors, 221 were for biological nutrient removal (BNR) facilities, 33 were from nitrification facilities, and 22 from organics removal facilities.

Table S20. Baseline emission factors for nitrous oxide based on facility type.

Category	N ₂ O-N [%]
Facility with nitrification	0.215
Facility with biological nutrient removal	0.439
Facility with organics removal only	0.146
Aerobic lagoon ¹⁹	1.6
Anaerobic lagoon ¹⁹	0
Facultative lagoon ¹⁹	0
Uncategorized lagoon	1.31

We also included process (non-combustion) CO₂ emissions, produced by biological processes onsite. The carbon content in wastewater consists of two components: the fraction produced from short-lived biogenic material (modern) and fraction derived from fossil-origin carbon. Recent studies on the fraction of fossil-origin carbon in wastewater are discussed in detail in **Supplementary Note 1**. In our analysis, we assumed that the baseline influent COD is 508 mg/L for all treatment trains and 11.9% of the influent COD is fossil-origin,²¹ the average of values reported in the literature, which range from 3-25%.²²⁻²⁴ We assumed a baseline of 54% of influent COD of fossil origin is assimilated into biomass and the rest released as CO₂.²³

2.5. Biosolids handling

2.5.1. Biosolids production and disposal

To estimate biosolid production, we used the EPA Biosolids Annual Report which contains biosolids generation and management practice information submitted electronically through the NPDES eReporting Tool by wastewater treatment plants and other industries.²⁵ Many facilities reporting to NPDES are not wastewater treatment plants (e.g. composting facilities, drinking water treatment plants) but may have names nearly identical to facilities in our wastewater treatment plant inventory. However, the dataset lacks a common but unique facility identification number (such as CWNS number) to directly match a facility in the Biosolids Report to the facilities in our

inventory. Thus, as a first step in data cleaning, we removed facilities listed in the Biosolids Report that likely were not wastewater treatment plants, the workflow for which is depicted in **Figure S14**, and summarized here.

First, we filtered the dataset using the standard industrial classification (SIC) codes associated with each NPDES permit, obtained from the national SIC code database maintained by U.S. EPA on facilities requiring environmental regulation. We kept all facilities that had SIC codes affiliated with sewer-systems (4952, 4953, and 4959). Next, we checked which facilities were listed as publicly owned treatment works (POTW) in the Reporting Obligation(s) column of biosolids report data. Because POTW may be either wastewater or drinking water facilities, we kept all facilities that were listed as POTW but did *not* also have a water supply SIC code (4941). Of the remaining facilities, we removed all facilities with SIC codes that make them unlikely to be municipally-owned wastewater treatment facilities (listed in **Table S21**, descriptions from the NAICS and SIC Crosswalk). We kept all facilities that did not have an SIC code match. Following these automated filtering steps, we manually checked publicly available online information on the remaining facilities. There were five facilities that required manual checks, summarized in **Table S22**, none of which were municipal wastewater treatment plants and thus were removed from the dataset.

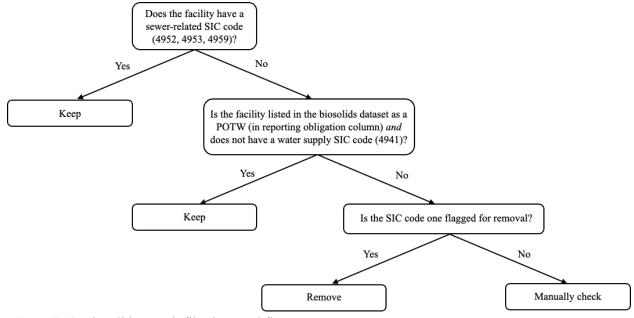


Figure S14. Biosolids permit filtering workflow.

Table S21. SIC codes associated with facilities in the biosolids dataset that were flagged for removal. A facility was removed from the Biosolids Dataset if it failed the first two exclusion criteria in **Figure S14**, and was associated with one of the SIC codes listed in this table.

SIC Code	Description	
1389	Oil & gas field services	

1522	Residential construction
2011	Meat packing plants
2491	Wood preserving
2493	Reconstituted wood products
2621	Paper mills
2899	Chemical preparation (spice/food extraction)
3171	Handbags & purses
3331	Primary copper
3498	Fabricated pipe & fitting
3533	Oil and gas field machinery
3743	Railroad equipment
4011	Railroads
4581	Airports
4911	Electric services
5075	Heating & cooling
5541	Gas station services
5812	Eating places
6514	Dwelling operators (residential)
6515	Mobile homes
6531	Real estate agents & managers
7011	Hotels & motels
7032	Sporting and recreation camps
7033	Trailer parks/campsites
7041	Membership hotels
7997	Sports/recreation clubs
7999	Amusement and recreation
8051	Skilled nursing care
8063	Psychiatric hospitals
8211	Schools
8221	Colleges & universities
8661	Religious organizations
9223	Correctional facilities
9711	National security

643 644 **Tab**

Table S22. List of facilities manually inspected for inclusion or exclusion from biosolids dataset. Note facility names appear as written in the downloaded EPA report.

Facility Name	SIC Code and Description	<u>Notes</u>	
Live Oak County Safety	7299 (misc. personal services)	Rest area waste facility – not a municipal	
Rest Area WWTF	7299 (misc. personal services)	wastewater treatment facility	
Bayou Club WWTP	8641 (civic & social association)	Wastewater treatment at a club – not a	
Bayou Club W W IF	8041 (CIVIC & SOCIAI ASSOCIATION)	municipal wastewater treatment facility	
CE Daalraged Derven Input	3511 (turbines/turbine generators)	General Electric is a private company – not	
GE Packaged Power Jport	7699 (repair services)	municipal wastewater treatment facility	

Sigmapro WWTP	6519 (real property lessors)	Sigma Pro is a private company - not municipal wastewater treatment facility
US DOE/Savannah River Site	2819 (industrial inorganic chemicals) 9611(administration of general economic programs)	US Department of Energy facility – not a municipal wastewater treatment plant

We then removed the top and bottom 10% of facilities from the updated EPA dataset, ranked by flow-weighted biosolids production rate (i.e., the ratio of biosolids produced to wastewater influent) as outliers. This resulted in a coverage of 2,124 facilities, leaving a large data gap in the amount and fate of biosolids at most treatment facilities in our inventory. Therefore, we supplemented the dataset by estimating biosolids production based on facility flow rate using **Equations S1-S3** from Seiple et al.²⁸ This approach assumes total dry sludge (M_T) is the sum of the dry sludge produced during primary treatment (M_P) and secondary treatment (M_S), as per **Equation S1**. For treatment trains without primary treatment, M_P is zero.

$$M_T = M_P + M_S$$
 Equation 1

For facilities with primary treatment, we used **Equation S2** to estimate the baseline M_P as a function of average influent flow rate to the facility (Q), total suspended solids (TSS), and the fraction of solids removed during primary settling (f). We used the average flow rate for each facility, as reported in CWNS. We assumed an average value of 260 mg/L for TSS, and an f value of $0.6.^{28}$

$$M_P = Q*TSS*f$$
 Equation 2

We used **Equation S3** to calculate the baseline total solids produced in secondary treatment. Here, biosolids production is again a function of flow rate (Q), as well as the biological oxygen demand entering the plant (BOD₅, or S₀ in Equation 3), assumed to be a standard concentration of 230 mg/L. The share of BOD₅ assumed to be converted into excess biomass is k, assumed to be 0.4, and f_v is the ratio between the fraction of TSS that is volatile suspended solids, assumed to be $0.85.^{28}$

$$M_S = Q \left[(k^*S_0) + \left(((1-f)^*TSS)^*(1-f_V) \right) \right]$$
 Equation 3

The estimated biosolids amount for those facilities is further adjusted if solid digestion is present. Specifically, we assumed a baseline volatile solids to total solids (VS/TS) ratio of 0.6 for produced solids and a baseline VS reduction ratio of 0.425 after anaerobic digestion and 0.475 after aerobic digestion. Note whenever multiple treatment trains are assigned to a facility, we assumed equal division of flow and calculated biosolids amount separately before adding them to get the total biosolids amount.

Next, we estimated the fate of captured biosolids for the facilities with a calculated biosolids amount. We first used biosolids disposal methods included in CWNS to identity the biosolids fate and, whenever multiple fates are indicated for one facility, we used ratios reflecting the distribution of biosolids disposal on a national level (**Table S23**) to split the biosolids.³¹

Table S23. Assumed breakdown of biosolids handling for different technologies.

		•	•	
Data set indicates:	Share of Biosolids			
Data set mulcates.	Landfill	Land Application	Incineration	
Landfill only	100%	0	0	
Land application only	0	100%	0	
Incineration only	0	0	100%	
Landfill and land application	28.6%	71.4%	0	
Land application and incineration	0	81.1%	18.9%	
Landfill and incineration	63.2%	0	36.8%	
Landfill, land application, and incineration	24.5%	61.2%	14.3%	

For the facilities where biosolids handling is not indicated through EPA's biosolids database or through CWNS, we estimated biosolids fate based on assigned treatment trains. For facilities with only one treatment train, we assumed all biosolids are incinerated if the treatment train includes incineration. Otherwise, we assumed a breakdown between landfilling and land application reflecting the distribution of biosolids disposal on a national level (**Table S23**). For plants with more than one treatment train, we first calculated biosolids produced from incineration trains using the methods mentioned above (**Equations 1-3**) based on the flow allocated to incineration trains, then the remaining of biosolids were split according to the national distribution between landfill and land application (**Table S23**). While land application may be more common in many places across the United States, states are increasingly banning land application of biosolids out of concerns of PFAS entering the food system. This changing regulatory context which will rapidly shift the distribution of biosolids fates, impacts resulting emissions.

2.5.2. Biosolids emission factors

We derived methane emission rate for biosolids sent to landfills using U.S. EPA's Landfill Gas Emissions Model (LandGEM). LandGEM estimates emission rates for total landfill gas, methane, carbon dioxide, non-methane organic compounds, and individual air pollutants from municipal solid waste (MSW) landfills³³. We used the first-order decomposition rate equation (**Equation S4**) to model baseline annual methane generation from landfills:

$$Q_{CH_4} = \sum_{i=1}^{n} \sum_{j=0.1}^{1} k L_0 \left(\frac{M_i}{10} \right) e^{-kt_{ij}}$$
 Equation 4

where:

- Q_{CH_4} is annual methane generation in the year of the calculation (m³/year),
- $\sum_{i=1}^{n}$ is summing over each year of waste accepted into the landfill (from year 1 to year n),
- $\sum_{i=0.1}^{1}$ is dividing each year into 10 increments of 0.1 years,
- k is methane generation rate constant (year⁻¹),

- L_0 is potential methane generation capacity (m³/megagram),
- M_i is the mass of waste accepted in the ith year (megagram), and
- t_{ij} is the age of the jth increment of waste M_i accepted in the ith year (decimal years, e.g., 3.2 years).

LandGEM calculates the CH₄ produced each year from the waste deposited (**Figure S15**). CH₄ generation decreases as waste decomposes, governed by the first-order decay constant (k) and the potential methane generation capacity (L_0). We used regulatory default values, according to Clean Air Act conventional: $k = 0.05 \text{ year}^{-1}$, $L_0 = 170 \text{ m}^3 \cdot \text{megagram}^{-1}$. These regulatory defaults were developed for compliance purposes and are therefore conservative values intended to protect human health. The density of CH₄ is assumed to be 0.68 kg/m³ at 15 °C and 1 atm³⁴.

The model results show that CH₄ generation is 8.312 m³/megagram (equivalent to 5.65 kg/tonne at 15 °C and 1 atm) in the first year after waste is deposited. Total CH₄ generation will reach 167.303 m³/megagram (equivalent to 113.766 kg/tonne at 15 °C and 1 atm) when summing all years since the waste was accepted into the landfill (from year 1 to year 80).

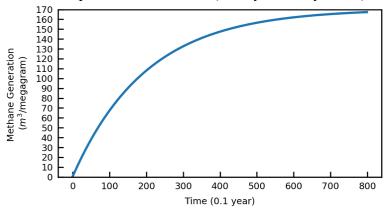


Figure S15. Cumulative methane emission from the biosolids landfill over 80 years.

CH₄ generation is highest in the first year and decreases as the waste decomposes over time. Since this study focuses on the short-term impact of emissions, we used CH₄ generation rate in the first year (5.652 kg/tonne at 15 °C and 1 atm) as the baseline CH₄ emission factor for analysis.

For biosolids used in land application, the N_2O is the primary greenhouse gas of concern.³⁵ We assumed a baseline of 4.9% nitrogen weight percentage in sludge and used the IPCC baseline emission factor of 0.01 kg N_2O -N/kg N in biosolids. Because carbon in biosolids is overwhelmingly biogenic,³⁶ we assumed all CO_2 from incineration is biogenic and therefore do not include it in our emissions estimates. The amounts of CH_4 and N_2O from incineration are also

739 minimal.

740 741

742

743

2.6. Uncertainty and sensitivity analyses

We used a tiered system modified from Feng et al. 2024 and 2025^{37,38} to determine the distribution and the range of uncertain parameters included in this study (**Table S24**). The criterion is listed below.

744745746

Criterion 1. For parameters with distributions in the literature, we used reported distributions as the literature.

747748749

750

751

Criterion 2. For parameters with ranges reported in literature, we chose a uniform distribution with the minimum and maximum limits as the lower and upper bounds of the distribution. If a widely accepted empirical value was also reported, we used it as the baseline; otherwise, we used the average of the minimum and maximum limits.

752753

754

755

Criterion 3. For parameters with only a widely accepted empirical value in the literature but no reported distribution, we assigned the accepted value as the baseline and assumed a uniform distributions with lower and upper bounds of 90% to 110% of the baseline value.

756 757

Criterion 4. For parameters with no known distribution in the literature:

758759

Criterion 4.1. If more than 20 data points were found in the literature, we selected triangular distributions and used 5th, 50th, and 95th percentiles from the literature values as the minimum, most probable, and maximum values.

760761762

763

Criterion 4.2. If only 5 to 20 data points were found in the literature, we selected triangular distributions and used minimum, average, and maximum of the literature value as the minimum, most probable, and maximum values.

764765766

Criterion 4.3. If less than 5 data points were found in the literature, we selected uniform distributions and used 80%, 100%, and 120% of the average value as lower bound, baseline, and upper bounds.

768769770

Table S24. List of parameters included in uncertainty and sensitivity analyses.

Parameters		Unit	Crit.	Dist.	Lower	Baseline	Upper	Ref.
CH ₄ global warming potential		kg CO ₂ -eq·kg CH ₄ -1	#3	uniform	26.8	29.8	32.8	16
N ₂ O global warn	ning potential	kg CO ₂ -eq·kg N ₂ O ⁻¹	#3	uniform	246	273	300	16
L-a L-n	kWh∙MG ⁻¹	#4.3	uniform	1110	1390	1660	39	
	L-n	kWh·MG ⁻¹	#4.3	uniform	528	660	792	39
electricity requirement	L-f	kWh·MG ⁻¹	#4.3	uniform	528	660	792	39
	L-u	kWh·MG ⁻¹	#4.3	uniform	1000	1260	1510	39
	others	kWh·MG ⁻¹	#2	uniform	treatment train-specific ^a		ific ^a	5
natural gas requirement kWh·MG ⁻¹ #2 uniform treatment train-specific		ific ^a	5					

influent COD		mg COD·L ⁻¹	#2	uniform	339	508	1016	13
CII : :	with AD	kg CH ₄ ·m ⁻³	#1	log-normal	shape = 0.60)1, scale =	0.00253	17
CH ₄ emission factor	without AD	kg CH ₄ ·m ⁻³	#1	Weibull	shape = 1.98	8, scale = 0	.000995	17
Tactor	L-n, L-f	kg CH ₄ ·kg ⁻¹ COD	#2	uniform	0	0.05	0.0975	18
influent TN		mg N·L ⁻¹	#2	uniform	23	35	69	13
	organics removal	kg N ₂ O-N·kg N ⁻³	#4.1	triangular	0.0000355	0.00146	0.0062	20
N ₂ O emission	nitrification	kg N ₂ O-N·kg N ⁻³	#4.1	triangular	0.000301	0.00215	0.0464	20
factor	BNR	kg N ₂ O-N·kg N ⁻³	#4.1	triangular	0.0000441	0.00439	0.0386	20
	L-a	kg N ₂ O-N·kg N ⁻³	#2	uniform	0.00016	0.016	0.045	18
influent fraction fossil origin		-	#4.2	triangular	0.001	0.119	0.279	21
fraction of assimilated COD of fossil origin		-	#4.3	uniform	0.428	0.535	0.642	23
electricity emission factor		kg CO ₂ -eq·kWh ⁻¹	#4.1	triangular	0.0102	0.355	0.899	15,40
natural gas	upstream	g CO ₂ -eq·MJ ⁻¹	#3	uniform	11.4	12.7	14.0	15
emission factor	combustion	g CO ₂ -eq·MJ ⁻¹	#3	uniform	50.7	56.3	61.9	15
sludge yield (pri secondary)	mary and	kg solids·m ⁻³	#3	uniform	0.238	0.264	0.290	28
sludge yield (see	condary only)	kg solids·m ⁻³	#3	uniform	0.118	0.131	0.144	28
Influent volatile fraction (VS/TS		-	#2	uniform	0.4	0.6	0.8	13,29,3
VS reduction	Anaerobic digestion	-	#2	uniform	0.35	0.425	0.5	13
fraction	Aerobic digestion	-	#2	uniform	0.4	0.475	0.55	13
CH ₄ emission factor during landfilling		kg CO ₂ -eq·tonne ⁻¹	#3	uniform	5.09	5.65	6.22	33
nitrogen mass fraction in land applied biosolids		-	#4.1	triangular	0.0122	0.049	0.062	41
nitrogen fraction emitted as N ₂ O during land application		-	#2	uniform	0.002	0.01	0.018	35

^a Values were obtained from the original model in Tarallo et al., which provides electricity and natural gas usage under both typical and best-practice scenarios. The lower of the two values (typical or best-practice) was set as the lower bound, and the higher value was set as the baseline. The upper bound was assumed to be equidistant from the baseline as the lower bound, but in the opposite direction. If the typical and the best-practice scenarios have the same value, no uncertainty was assumed.

For CH₄ emission factors, we chose the best-fit distribution from three types of distribution mentioned in the original source (Weibull distribution, Gamma distribution, log-normal distribution).¹⁷ The fitness was measured by Kolmogorov-Smirnov test, with a higher p-value and a lower test statistic indicating higher fitness (**Table S25** and **Table S26**).

Table S25. Fitness of different distributions on the CH₄ emission factor data for treatment configurations with anaerobic digestion.

Distribution	p-value	Kolmogorov-Smirnov test statistic	Best-fit
Weibull	0.796	0.191	No
Gamma	0.909	0.165	No
log-normal	0.966	0.145	Yes

Table S26. Fitness of different distributions on the CH₄ emission factor data for treatment configurations
 without anaerobic digestion.

Distribution	p-value	Kolmogorov-Smirnov test statistic	Best-fit
Weibull	0.845	0.141	Yes
Gamma	0.565	0.182	No
log-normal	0.812	0.146	No

2.6.1. Uncertainty values reported in main text

The error bars and uncertainty values depicted in figures and tables of the main text reflect the results of a few different types of analysis. While values are mentioned in the main text figures and captions, for clarity here we provide full details here as well.

- **Figure 2b**: electricity bounds reflect data from all 134 regions for which we simulated electricity mix. Baseline value (bar height) represents the median value of a uniform distribution, with error bars representing the 5th and 95th percentiles. We used the national emissions profile in order to demonstrate the potential variability inherent in location of a facility, as opposed to using the electricity carbon intensity associated with the actual plants in our dataset (used in later calculations).
- **Figures 2c&d**: boxplots reflect results of the Monte Carlo simulation with the electricity grid carbon intensity using the national electricity values same as **Figure 2b**.
- **Figures 4&5**: the facility-level total emission were approximated as the sum of median values of different emission types, with the electricity-related emissions calculated using the regional electricity mix corresponding to each specific facility.
- Table 1: the uncertainty of national-level emissions related to electricity was calculated with the regional electricity-mix data as the baseline and by assuming a uniform distribution with 80% and 120% of the baseline value as the lower and upper bound, respectively. The uncertainty of nation-level emissions related to biosolids disposal was calculated using the national total biosolids mass flow estimated by summing the facility-level data (see Section 2.5) as the baseline and by assuming a uniform distribution with 80% and 120% of the baseline value as the lower and upper bound, respectively.

2.7. Data access

Publicly available data used in this study was available freely online at the time of publication. We created Permalink archives of access URLs in order to ensure future data availability, as summarized in **Table S27**.

816 817

812

813

Table S27. Data access URLs and archived access links.

Data Source	Reference	Access URL at time of publication	URL Archive
U.S. Environmental Protection Agency's Inventory of U.S. Greenhouse Gas	Main text - 7	https://www.epa.gov/ghgemissions/inventor y-us-greenhouse-gas-emissions-and-sinks- 1990-2021	Access website: https://perma.cc/V W7K-N8T9
Emissions and Sinks a second time			PDF archive: https://perma.cc/Y6 WT-HUGP
NREL Standard Scenarios Cambium Data for 2020	Main text - 17	Data downloader: https://scenarioviewer.nrel.gov/?project=57 9698fe-5a38-4d7c-8611- d0c5969b2e54&layout=Default%20Layout &mode=view Data visualizer: https://scenarioviewer.nrel.gov/?project=57 9698fe-5a38-4d7c-8611- d0c5969b2e54&layout=Default%20Layout	Data visualizer archive: https://perma.cc/3K 97-CJDY
Life Cycle and Cost Assessments of Nutrient Removal Technologies in Wastewater Treatment Plants	Main text - 20	https://www.epa.gov/system/files/document s/2023-06/life-cycle-nutrient-removal.pdf	Website archive: https://perma.cc/E X52-MEC5
U.S. Environmental Protection Agency's Biosolids Biennial Report No. 9	Main text - 40	https://www.epa.gov/system/files/document s/2022-12/2020-2021-biennial-report.pdf	Website archive: https://perma.cc/76 FR-PYPQ
NREL Regional Energy Deployment System	Main text - 44	Model website: https://www.nrel.gov/analysis/reeds/index GitHub link with shape files used for generating regional energy mix used in this study: https://github.com/NREL/ReEDS-2.0/tree/main/inputs/shapefiles/US_PCA	Model website archive: https://perma.cc/E U47-2BEJ GitHub archive: https://perma.cc/N GM2-737X
U.S. Environmental Protection Agency's overview of national biosolids disposal	Supplementary information - 25	https://www.epa.gov/biosolids/basic- information-about-sewage-sludge-and- biosolids	Website archive: https://perma.cc/RJ U5-X37R

3. Supplementary Note 1: fossil origin carbon in wastewater

GHG emission guidelines traditionally focus on CH₄ and/or N₂O emissions, excluding CO₂ emissions arising from biological treatment processes. This exclusion is based on the argument that such emissions primarily originate from biogenic organic matter in human excreta or food waste, and therefore, they are typically not accounted for in national total emissions.⁴² Nevertheless, wastewater treatment plants also emit fossil CO₂ and relying only on the assumption that all on-site CO₂ emissions are biogenic may lead to an underestimation of GHG emissions. The IPCC 2006 Guidelines established an international convention that discourages the reporting of non-biogenic CO₂ from activities in the waste sector. However, IPCC 2019 acknowledges the need for future improvements to the IPCC Guidelines, including a method for estimating non-biogenic emissions associated with wastewater treatment operations and wastewater discharges.¹⁸ The proportions of biogenic and fossil carbon fractions depend on various factors, including wastewater characteristics and type. In many cases, accurate data regarding the origin of waste is either unavailable or outdated.

Griffith et al.²² reported that 25% of the total organic carbon (TOC) in wastewater originates from fossil sources, likely derived from cleaning products, pharmaceuticals, and fossil-fuel-based items. Another study conducted by Law et al.²³ suggested that existing GHG accounting guidelines, which assume that all CO₂ emissions from wastewater are biogenic, may lead to an underestimation of emissions. They conducted radiocarbon isotopes research and found that 4-14% of TOC in wastewater is of fossil origin. Additionally, Liu et al.²⁴ indicated that fossil carbon constitutes 3-10% of the total carbon in the mixed sludge. Their study revealed the effect of digestion on the removal of fossil carbon from wastewater as well as the CO₂ emissions from wastewater. Wang et al.² constructed an emission inventory of wastewater treatment facilities for CH₄, N₂O and CO₂ emissions from different treatment processes, energy consumptions, and effluent discharge for the time-period from 2006 to 2019 in China. However, the study did not distinguish between fossil CO₂ and biogenic CO₂ emissions from biological treatment but regarded CO₂ emission as the sum of fossil CO₂ and biogenic CO₂ emissions. Additional widespread measurements of CO₂ emissions at wastewater treatment plants that quantifies the portion that is of fossil carbon will improve wastewater treatment plant greenhouse gas inventories.

849 References

- 1. Parravicini, V., Nielsen, P. H., Thornberg, D. & Pistocchi, A. Evaluation of greenhouse gas
- emissions from the European urban wastewater sector, and options for their reduction. Sci.
- 853 *Total Environ.* **838**, 156322 (2022).
- 2. Wang, D. et al. Greenhouse gas emissions from municipal wastewater treatment facilities in
- 855 China from 2006 to 2019. *Sci. Data* **9**, 317 (2022).
- 3. Chen, W., Zhang, Q., Hu, L., Geng, Y. & Liu, C. Understanding the greenhouse gas emissions
- from China's wastewater treatment plants: Based on life cycle assessment coupled with
- statistical data. *Ecotoxicol. Environ. Saf.* **259**, 115007 (2023).
- 4. U.S. Environmental Protection Agency. *Inventory of U.S. Greenhouse Gas Emissions and*
- Sinks 1990-2022. https://www.epa.gov/ghgemissions/inventory-us-greenhouse-gas-emissions-
- and-sinks-1990-2022 (2023).
- 5. Tarallo, S., Shaw, A., Kohl, P. & Eschborn, R. A Guide to Net-Zero Energy Solutions for
- Water Resource Recovery Facilities. https://iwaponline.com/ebooks/book/293/ (2015).
- 6. U.S. Environmental Protection Agency. Clean Watersheds Needs Survey 2004. (2008).
- 7. U.S. Environmental Protection Agency. Clean Watersheds Needs Survey 2008. (2010).
- 8. U.S. Environmental Protection Agency. Clean Watersheds Needs Survey 2012. (2016).
- 9. U.S. Environmental Protection Agency. Clean Watersheds Needs Survey 2022. (2024).
- 10. U.S. Department of Energy (DOE). U.S. Department of Energy Combined Heat and Power
- 869 Installation Database. (2024).
- 870 11. Water Environment Federation (WEF). Water Resource Recovery Facilities Biogas Data.
- 871 (2024).
- 12. U.S. Environmental Protection Agency. Lagoon Inventory Dataset. (2022).

- 13. Tchobanoglous, G. et al. Chapter 13: Processing and Treatment of Sludges. in Wastewater
- 874 Engineering: Treatment and Resource Recovery (McGraw-Hill Education, 2014).
- 875 14. United States Environmental Protection Agency. *Biosolids Technology Fact Sheet: Alkaline*
- 876 Stabilization of Biosolids. https://www.epa.gov/sites/default/files/2018-
- 877 11/documents/alkaline-stabilization-biosolids-factsheet.pdf (2000).
- 878 15. Wang, M. et al. Greenhouse gases, Regulated Emissions, and Energy use in Technologies
- 879 Model ® (2023 Excel). https://doi.org/10.11578/GREET-Excel-2023/dc.20230907.1 (2023).
- 880 16. IPCC. Climate Change 2022: Impacts, Adaptation, and Vulnerability. Contribution of
- Working Group II to the Sixth Assessment Report of the Intergovernmental Panel on Climate
- 882 *Change*. https://www.ipcc.ch/report/ar6/wg2/about/how-to-cite-this-report/ (2022)
- 883 doi:10.1017/9781009325844.
- 17. Song, C. *et al.* Methane Emissions from Municipal Wastewater Collection and Treatment
- 885 Systems. *Environ. Sci. Technol.* **57**, 2248–2261 (2023).
- 18. Bartram, D. et al. Wastewater Treatment and Discharge. in 2019 Refinement to the 2006
- 887 IPCC Guidelines for National Greenhouse Gas Inventories (Intergovernmental Panel on
- Climate Change, 2019).
- 19. Barton, L. et al. Sampling frequency affects estimates of annual nitrous oxide fluxes. Sci.
- 890 *Rep.* **5**, 15912 (2015).
- 891 20. Song, C. et al. Oversimplification and misestimation of nitrous oxide emissions from
- wastewater treatment plants. *Nat. Sustain.* (2024) doi:10.1038/s41893-024-01420-9.
- 21. Tseng, L. Y. et al. Identification of Preferential Paths of Fossil Carbon within Water
- Resource Recovery Facilities via Radiocarbon Analysis. *Environ. Sci. Technol.* **50**, 12166–
- 895 12178 (2016).

- 896 22. Griffith, D. R., Barnes, R. T. & Raymond, P. A. Inputs of Fossil Carbon from Wastewater
- Treatment Plants to U.S. Rivers and Oceans. *Environ. Sci. Technol.* **43**, 5647–5651 (2009).
- 23. Law, Y., Jacobsen, G. E., Smith, A. M., Yuan, Z. & Lant, P. Fossil organic carbon in
- wastewater and its fate in treatment plants. *Water Res.* 47, 5270–5281 (2013).
- 900 24. Liu Chen, Oshita Kazuyuki, Takaoka Masaki, & Fukutani Satoshi. Behaviour of Fossil and
- 901 Biogenic Carbon in Sewage Sludge Treatment Processes and Their Impacts on Greenhouse
- 902 Gas Emissions. *Chem. Eng. Trans.* **89**, 97–102 (2021).
- 903 25. U.S. Environmental Protection Agency. Biosolids Biennial Report No. 9 (Reporting Period
- 904 *2020-2021*). https://www.epa.gov/system/files/documents/2022-12/2020-2021-biennial-
- 905 report.pdf (2022).
- 906 26. United States Environmental Protection Agency. Facility Registry System.
- 907 27. NAICS Association. NAICS & SIC Crosswalk. https://www.naics.com/product/sic-naics-
- 908 cross-references/ (2023).
- 909 28. Seiple, T. E., Coleman, A. M. & Skaggs, R. L. Municipal wastewater sludge as a sustainable
- bioresource in the United States. J. Environ. Manage. 197, 673–680 (2017).
- 911 29. Tchobanoglous, G. et al. Chapter 3: Wastewater Flowrates and Constituent Loadings. in
- Wastewater Engineering: Treatment and Resource Recovery (McGraw-Hill Education, 2014).
- 913 30. Selivanovskaya, S. Yu., Zaripova, S. K., Latypova, V. Z. & Hung, Y.-T. Treatment and
- Disposal of Biosolids. in *Environmental Bioengineering* (eds. Wang, L. K., Tay, J.-H., Tay, S.
- 915 T. L. & Hung, Y.-T.) (Humana Press, Totowa, NJ, 2010). doi:10.1007/978-1-60327-031-1.
- 916 31. United States Environmental Protection Agency. Basic Information about Biosolids.
- 917 *Biosolids* https://www.epa.gov/biosolids/basic-information-about-biosolids (2023).

- 918 32. Pozzebon, E. A. & Seifert, L. Emerging environmental health risks associated with the land
- application of biosolids: a scoping review. *Environ. Health* **22**, 57 (2023).
- 920 33. U.S. Environmental Protection Agency. Landfill Gas Emissions Model (LandGEM). (2023).
- 921 34. Methane | Gas Encyclopedia Air Liquide. https://encyclopedia.airliquide.com/methane.
- 922 35. Intergovernmental Panel on Climate Change. Chapter 11. N2O Emissions from Managed
- 923 *Soils, and CO2 Emissions from Lime and Urea Application.* (2019).
- 36. Intergovernmental Panel on Climate Change. Chapter 5. Incineration and Open Burning of
- 925 Waste. (2019).
- 926 37. Feng, J., Li, Y., Strathmann, T. J. & Guest, J. S. Characterizing the Opportunity Space for
- 927 Sustainable Hydrothermal Valorization of Wet Organic Wastes. *Environ. Sci. Technol.* **58**,
- 928 2528–2541 (2024).
- 929 38. Feng, J., Strathmann, T. & Guest, J. Financially Driven Hydrothermal-based Wastewater
- Solids Management for Targeted Resource Recovery and Decarbonization in the Contiguous
- 931 U.S. Preprint at https://doi.org/10.26434/chemrxiv-2025-qfxwd (2025).
- 932 39. U.S. Environmental Protection Agency. Principles of Design and Operations of Wastewater
- 933 Treatment Pond Systems for Plant Operators, Engineers, and Managers.
- https://www.epa.gov/sites/default/files/2014-09/documents/lagoon-pond-treatment-2011.pdf
- 935 (2011).
- 40. Gagnon, P., Frazier, W., Hale, E. & Cole, W. Cambium data for 2020 Standard Scenarios.
- National Renewable Energy Laboratory (2020).
- 938 41. Rigby, H. et al. A critical review of nitrogen mineralization in biosolids-amended soil, the
- associated fertilizer value for crop production and potential for emissions to the environment.
- 940 *Sci. Total Environ.* **541**, 1310–1338 (2016).

- 941 42. Intergovernmental Panel on Climate Change. Chapter 6. Wastewater Treatment and
- 942 *Discharge*. (2019).