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Supporting Information for:

Assessing the Energy and Environmental Performance of Algae-Mediated Tertiary Treatment of Estrogenic Compounds

Lisa M. Colosi, Eleazer P. Resurreccion, Yongli Zhang

24 pages, 12 tables, and 4 figures

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1 Municipal Wastewater Treatment and System Water Balances

1.1 Overview and Assumed Effluent Quality

Four hypothetical wastewater treatment plant (WWTP) systems were modeled using LCA, one each corresponding to four selected tertiary treatments systems: algae, ozonation (OZ), UV irradiation (UV), and sorption onto granular activated carbon (GAC). The selected systems were sized to deliver one functional unit (FU), which corresponds to treatment of 10 million gallons per day (MGD) over one year. Literature data for electricity consumption at municipal WWTPs was based primarily on Menendez (2010), with supplemental information corresponding to the Moore's Creek WWTP in Charlottesville, Virginia. All material and energy inputs required to deliver one FU in each system were accounted for, but infrastructure burdens (for capital equipment, construction, etc) were not included.

Table S1 summarizes permit levels for pertinent wastewater constituents in the final discharged effluents. It was assumed that all four tertiary treatments, and the reference "WWTP" case (no tertiary treatment), are required to produce effluent that is compliant with these specifications. It was also assumed that all primary and secondary treatment operations are the same across all five evaluated systems.

Source					
, 2013					
, 2007					
2010)					
2010)					
, 2005					

Table S1. Final effluent levels for selected wastewater constituents.

1.2 Influent Collection, Pretreatment, and Primary Treatment

Electricity consumption corresponding to influent collection pretreatment, and primary treatment was based on data collected by Menendez (2010) for a typical 10-MGD municipal WWTP in the Southeastern USA. Pumping electricity demand is 1,402 kWh/d (Menendez, 2010). Screening accounts for 2 kWh/d, and aerated grit removal consumes 134 kWh/d (Menendez, 2010). Operation of primary clarifiers accounts for another 155 kWh/d (Menendez, 2010). An additional 800 kWh/d is consumed for lighting and in buildings on site (Menendez, 2010). All of these values were converted to MJ and then multiplied by 365 days per year to compute electricity consumption in MJ/y.

Retention time during primary clarification was assigned to a triangular distribution, with min = 2 h, likeliest value = 2.5 h, and max = 3 h (Tchobanoglous et al, 2003). The BOD concentration flowing into primary treatment was assigned to a uniform distribution, based on typical municipal wastewater: min = 120 mg/L, max = 380 mg/L (Tchobanoglous et al, 2003). BOD removal efficiency during primary treatment was computed using Equation S1, where *HRT* is hydraulic retention time (days), *a* = 0.018, and *b* = 0.020 (Tchobanoglous et al, 2003). The resulting value for this calculation was R_{BOD} = 36%.

Equation S1
$$R_{BOD} = \frac{HRT}{a+b \times HRT}$$

The TSS concentration flowing into primary treatment was also assigned to a uniform distribution, based on municipal wastewater: min = 120 mg/L, max = 370 mg/L (Tchobanoglous et al, 2003). TSS removal efficiency during primary treatment was computed using Equation S2, where a = 0.0075, and b = 0.014(Tchobanoglous et al, 2003). The resulting value for this calculation is R_{TSS} = 58%.

Equation S2
$$R_{TSS} = \frac{HRT}{a+b \times HRT}$$

TSS removal during primary treatment gives rise to primary solids, which are collected from the bottom of the clarifier and transported to anaerobic digestion. Typical concentrations of the primary solids stream are min = 50,000, likeliest = 60,000, and max = 90,000 mg/L (Tchobanoglous et al, 2003).

1.3 Secondary Treatment and P Removal

Secondary treatment comprises "advanced treatment with nitrification" (i.e., "biological nutrient removal – "BNR") plus methanol-dosed denitrification (Carey and Migliaccio, 2009; Menendez, 2010; RWSA, 2007). This combination of operations delivers TN concentrations on the range 3-8 mg/L and TP concentrations on the range 1-2 mg/L (Carey and Migliaccio, 2009). Alum precipitation is used to deliver phosphorus removal down to the desired effluent limit (0.1 mg/L). Table S10 summarizes pertinent characteristics of the secondary effluent produced using this system.

Table S2. Select	ed characteristics	s of the secondary	<pre>/ effluent produced</pre>	via advanced	biological	nitrogen
removal.						

Constituent	Distribution ^a	Source
BOD	Tri (6, 7, 12) mg/L	Barnard, 1975; Kim, 2009
TSS	U (13, 17) mg/L	Barnard, 1975; Kim, 2009
TN	U (3, 8) mg/L	Carey and Migliaccio, 2007
ТР	U (1, 2) mg/L ^b	Carey and Migliaccio, 2007; RWSA, 2007
Fecal coliform (MPN)	Tri (3.3E4, 7.9E4, 4.9E6) MPN/100 mL	Lin et al, 1973; Tyrrell et al., 1995
Alkalinity	U (50, 100) mg/L as CaCO $_3$	US EPA, 2008
рН	U (6.4, 7.2)	RWSA, 2007
17α -Ethynylestradiol (EE2)	Tri (0.2, 3.9, 7.5) ng/L	Baronti 2000; Cargouet 2004; Clara, 2005; Desbrow, 1998; Drewes, 2005;
17β-Estradiol (E2)	Tri (0.2, 8.6, 17) ng/L	and Sumpter, 2001; Joss, 2004; Huang and Sedlak, 2001; Leusch, 2005; Nakada, 2007: Ondo. 2002: Pobort. 2007; Spyder
Estrone (E1)	Tri (0.75, 24.9, 49.1) ng/L	1999; Tan, 2006; Ternes, 1999

^a Triangular distributions are denoted "Tri (min, likeliest, max)". Uniform distributions are denoted "U (min, max)". ^b Alum precipitation is used to achieve TP concentrations at or below the permitted level (0.1 mg/L). Electricity consumption is 5,320 kWh/d for aeration to oxidize carbonaceous BOD plus 3,446 kWh/d for biological nitrification (Menendez, 2010). A stream of volatile suspended solids (VSS) and fixed suspended solids (FSS) is produced during secondary treatment, whereby VSS + FSS = TSS. The VSS comprises aerobic heterotroph microorganisms, which consume the BOD flowing into secondary treatment and, as a result, grow and multiply. The quantity of VSS produced during secondary treatment was computed based on the mass of BOD removed during secondary treatment (i.e., BOD mass in the secondary influent minus BOD mass in the secondary effluent) multiplied by an empirical yield factor (Y). Typical values for Y are min = 0.3, likeliest = 0.4, and max = 0.5; units are "g VSS produced per g BOD consumed" (Tchobanoglous et al, 2003). It was assumed that only 50% of the BOD in the primary effluent (secondary effluent) is soluble and therefore biodegradable to form VSS.

FSS corresponds to the difference in TSS concentrations (i.e., TSS in secondary influent minus TSS in secondary effluent) multiplied by the total flow of primary effluent (i.e., secondary influent). VSS and FSS were added together to compute the total mass of secondary solids flowing from the secondary clarifiers into anaerobic digestion. The solids concentration in the secondary solids stream was assigned to a uniform distribution: min = 8,000 mg/L and max = 12,000 mg/L. Pumping of recycled activated sludge VSS (RAS) and "wasted" activated sludge VSS (WAS) consumes 85 kWh/d (Menendez, 2010). We did not account for CO_2 production (or other GWP effects) arising from production of VSS during secondary treatment.

An additional 155 kWh/d is consumed for operation of the secondary clarifiers (Menendez, 2010). Methanol consumption for denitrification was computed based on estimated extent of nitrification and literature values for methanol demand. Regarding extent of nitrification, it was assumed that no nitrate exists in the secondary influent and that all nitrogen exists as ammonia. The fraction of nitrogen converted into nitrate (i.e., extent of nitrification) was assigned to a triangular distribution based on data for typical NO₃-N and NH₄-N concentrations in secondary effluent from BNR WWTPs: min = 25%, likeliest = 70%, max = 88% (Carey and Migliaccio, 2009). Methanol demand for conversion of nitrate into N_2 gas (i.e., denitrification) was assigned to a uniform distribution over the range 3.4-8.9 kg methanol per 1 kg of NO₃-N denitrified (RWSA, 2007; Water Environmental Federation, 2009). Alum consumption is 5.5 kg $Al_2(SO_4)_3$ ("alum") for removal of 1 kg P, based on a 1:1 molar ratio of Al added to P removed (RWSA, 2007). Menendez (2010) also reports 2,022 kWh/d for dissolved air flotation and 508 kW/d for return and waste activated sludge pumping. Three additional parameters related to alum precipitationbased TP removal: 552 kWh/d for chemical addition, 822 kWh/d for filter feed pumping, and 385 kWh/d for filtration. For tertiary treatments capable of reducing the TN and/or TP concentrations beyond what was present in the secondary effluent, upstream burdens and electricity consumption corresponding to denitrification and alum precipitation procedures were scaled back from these baseline values. Table S11 summarizes information on life cycle impacts of methanol, alum, and other material inputs.

1.4 System Water Balances

Table S3 summarizes two types of water balances, corresponding to primary and secondary treatment plus either a conventional tertiary treatment (OZ, UV, or GAC) or algae-based conventional treatment. These water balances were used throughout the LCA modeling process to estimate quantities of materials flowing into and out of each unit operation.

Table S3. Water balances for municipal wastewater treatment coupled with either "conventional" tertiary treatment or "algae-mediated" tertiary treatment. All flows correspond to 1 FU, which is treatment of 10 MGD over one year.

Mass Flow (L/y)	Conventional	Algae-Mediated	Notes
Raw influent	13,816,755,750	13,816,755,750	10 MGD × 365 days/year
Primary solids	28,652,498	28,652,498	
Primary effluent	13,788,103,252	13,788,103,252	
Secondary solids	127,344,246	127,344,246	
Secondary effluent	13,660,759,007	13,660,759,007	
Evaporation from algae ponds	NA	61,574,178	CA conditions, Clarens et al (2010)
Algae slurry from ponds to AF ^a	NA	13,599,184,829	
Algae slurry post-AF/pre-TH ^a	NA	1,102,636,608	
Algae slurry post-TH/pre-AD ^a	NA	100,239,692	
Combined AD inlet	NA	256,236,435	
Dewatered AD sludge post-BFP ^a	9,863,371	11,595,927	
Tertiary effluent	13,660,759,007	13,498,945,137	

^a AF = auto-flocculation, TH = gravity thickening, AD = anaerobic digestion, BFP = belt-filter press.

For the water balances summarized in Table S3, the percent influent unaccounted for is 1.6% in the conventional systems and 1.7% in the algae system. Thus, the mass balances are mostly closed. Internal recycle within the systems boundaries was not explicitly accounted for in the mass balance.

2 Analysis of Algae Cultivation System as a Tertiary Treatment Step

2.1 Model Overview

An LCA model framework was used to analyze algae cultivation in open ponds as a possible tertiary treatment. This model was based on previous LCA studies of raceway pond systems (Clarens et al, 2011; Resurreccion et al, 2012). Figure S1 shows an overview of the proposed system, whereby: algae biomass from open pond reactors is subject to *autoflocculation* and gravity *thickening* for dewatering and then fed into *anaerobic digestion* with the primary and secondary sludges. The post-digestion slurry from the digester is dewatered using a *belt-filter press (BFP)*. The nutrient-rich liquid digestate is recycled back into primary treatment, and the biogas generated in anaerobic digestion is *combusted* to generate bioelectricity.



Figure S1. Process flow for a WWTP employing algae cultivation as tertiary treatment. The heavy dashed red box represents the system boundaries.

2.2 Sizing the Algae-Based Tertiary Treatment System

The sizing of the algae cultivation pond was based on HRT, however, two different HRTs were considered: (1) the HRT required to achieve "acceptable" estrogen removal via algae-mediated reactions in the cultivation ponds, whereby longer residence times yield better removal; and (2) the HRT yielding best algae harvest, because algae concentration increases up to some optimum residence time and then decreases thereafter. Based on these constraints, algae pond HRT was assigned to a triangular distribution: min = 0.5 day, max = 1.5 days, and likeliest value = 1 day. Corresponding pond volume and land use were roughly 59,000 m³ and 11.9 ha, respectively, for an assumed pond depth of 0.5 m (Benemann and Owald, 1996).

Estrogen removal for the range of algae pond HRTs evaluated in this study varies for individual estrogens: 64-80% for 17 β -estradiol (E2), 30-45% for estrone (E1), and 20-47% for 17 α -ethinylestradiol (E2). Section 4 summarizes additional information pertaining to estrogens.

2.3 Algae Cultivation and Conversion Processes

<u>Yield</u>. Algae yield was assigned to the uniform distribution 47.1-65.7 Mg/ha-yr. The minimum value corresponds to the likeliest yield of freshwater algae in very sunny climates (e.g., central California) from Clarens et al (2010); assuming the triangular distribution of radiation use efficiency is min = 0.6, likeliest = 2.1, and max = 2.8 g of algae dry solids per MJ of photosynthetically-active radiation (PAR), where PAR

varies from 5.0 (January) to 11.2 (July) MJ/m² (NREL, 1994). The maximum value corresponds to the projected yield for cultivation of freshwater algae in municipal wastewater effluent according to Menger-Krug et al (2012).

<u>Nutrient Procurement.</u> Required quantities of carbon (from carbon dioxide – CO_2), nitrogen, and phosphorus were computed stoichiometrically based on an assumed molecular composition given by $C_{106}H_{181}O_{45}N_{15}P$ (Clarens et al, 2010; 2011). For the range of yields specified above, there was no need to procure CO_2 , N, or P beyond what is recycled from other processes.

For CO_2 , it was assumed that the efficiency of uptake by the algae is log-normally distributed with mean = 0.7 and standard deviation = 0.1. Even so, the combined amount returned from anaerobic digestion and methane combustion was significantly greater than the amount required. This difference was accounted for as "direct" CO_2 contributions to global warming potential (GWP). In contrast, the CO_2 taken up by the algae via photosynthesis, then later digested to produce methane which is ultimately combusted to produce electricity, was not accounted for as a direct contribution to GWP, because it was assumed that the amount consumed is equal to the amount released.

For nitrogen and phosphorus, the concentrations in Table S2 were multiplied by the mass flow of secondary effluent per year (Table S3) to compute mass loading of each nutrient into the algae ponds. It was assumed that nitrification would be unchanged compared to the "just WWT" case, since nitrate is a desirable form of nitrogen for algae growth. In contrast, it was assumed that downstream TN and TP uptake by the algae could offset the need for some denitrification and alum trimming, respectively. For the <u>average</u> effluent nutrient concentrations, mass flows, and algae yields used in this study, roughly 50% of the available nitrogen (as nitrate) and 30% of the available phosphorus in the BNR effluent were taken up by the algae. The <u>average</u> residual nitrogen concentration, once allocated over the total mass flow of tertiary effluent, was sufficiently low to meet the TN limit in Table S1. In contrast, the residual phosphorus always required trimming, via alum dosing, to meet the TP permit level.

<u>Cultivation Mixing and Pumping</u>. It was assumed that paddle-wheel mixers are used to provide continuous mixing in the open ponds. Electricity consumption per paddlewheel was assigned to a triangular distribution (min = 0.0001, likeliest = 0.001, max = 0.01 kW), and it was assumed that ten paddle-wheels are required per ha of algae pond (Benemann and Oswald, 1996). It was also assumed that roughly 10 m of head is required to circulate the algae slurry out of the ponds and through subsequent conversion processes (Stephenson et al, 2011). The efficiency of the pumps required to provide this head was set to 85%.

<u>Algae Dewatering</u>. Because anaerobic digestion does not require biomass to be very dry, only autoflocculation (AF) and gravity thickening (TH) were used to provide bulk dewatering for the harvested algae. The concentration factor for AF was assigned to a triangular distribution: min = 5, likeliest = 10, and max = 22. It was assumed that the alum dosing for phosphorus trimming occurs downstream of AF, such that there is still sufficient phosphate in the solution to facilitate AF (Spilling et al, 2011). The concentration factor for TH was also assigned to a triangular distribution: min = 8, likeliest = 10, and max = 15. Electricity consumption for TH was 209 MJ per Mg of algae dry solids, from Soda et al (2010). Finally, it was assumed that the efficiency of the TH process is less than 100%, whereby not all of the algae biomass is captured and removed from the decanted liquid. The capture efficiency was assigned to a uniform distribution over the range 0.88-0.99 (Menger-Krug et al, 2012). <u>Anaerobic Co-digestion</u>. It was assumed that primary sludge, secondary sludge, and dewatered algae are co-digested together to produce biogas. This biogas is then combusted to produce bio-electricity, analogous to Case A of Clarens et al (2011). Table S4 summarizes pertinent information regarding the digestion of primary solids, secondary solids, and algae biomass. The assumed HRT for anaerobic digestion was roughly 20-40 days.

Table S4. Parameters related to anaerobic co-digestion of primary and secondary WWTP solids with the harvested algae biomass.^a

Parameter	Primary Solids	Secondary Solids	Algae Biomass
Solids concentration (mg/L)	66,667	10,000	4,260
Ash content (fraction)	U (0.27, 0.33)	U (0.12, 0.41)	Tri (0.02, 0.05, 0.07) ^b
VSS digestibility ("destruction")	U (0.56, 0.60)	U (0.56, 0.60)	Tri (0.41, 0.54, 0.68) ^c
Methane yield (L CH ₄ /g VSS digested)	U (0.75, 1.12)	U (0.75, 1.12)	Tri (0.22, 0.49, 0.54) ^d

a Triangular distributions are denoted "Tri (min, likeliest, max)". Uniform distributions are denoted "U (min, max)".
All values are from Tchobanoglous (2003) unless otherwise noted.

^b Algae ash content is from Resurreccion et al (2012).

^c Based on Golueke (1956), Cecchi et al (1996), and Yuan (2012).

^d Based on Cecchi et al (1996), Clarens et al (2012), Golueke et al (1956), and Park et al (2012)

Electricity and heat demands for operation of the anaerobic digesters were taken from Soda et al (2010). Electricity consumption was 288 MJ per Mg of dry algae solids digested. Heat demand was 907 MJ per Mg of dry algae solids digested.

Table S5 summarizes pertinent information regarding ancillary digestion operations, including: conversion of digestion biogas into electricity, recycling of biogas and methane-combustion CO₂ back to the algae cultivation ponds, and belt-filter pressing to dewater the digestate solids.

Parameter	Value	Source
Methane density at 25 °C (kg/m ³)	0.668	Clarens et al, 2011
Methane lower heating value (kJ/m ³)	35,800	Tchobanglous, 2003
Turbine efficiency for conversion of methane to electricity	0.65	Clarens et al, 2011
Biogas methane fraction (v/v)	Tri (0.46, 0.72, 0.76)	Sialve et al, 2009
CO ₂ yield from methane combustion (kg/kg)	2.8	Stoichiometry
Pumping energy for CO ₂ recycling back (J/kg CO ₂)	314	Clarens et al, 2011
BFP electricity consumption (MJ per kg solids)	356	Soda et al (2010)
Solids concentration of the BPF-dewatered solids (g/L) ^b	Tri (214, 235, 268)	Tchobanglous, 2003

Table S5. Parameters related to ancillary digestion operations.

^a It was assumed that biogas comprises only CO₂ + methane. ^b Specific gravity of the BFP-dewatered "cake" is 1.07.

2.4 Tertiary Effluent Quality for the Algae-Based Treatment

The "treated" effluent from the algae-based system comprises the decanted liquid from the autoflocculation and thickening steps. This is the effluent that is discharged from the WWTP; therefore, it must meet the effluent standards summarized in Table S1. It is currently unknown how algae-mediated tertiary treatment will affect the concentration of wastewater microorganisms, so to be conservative, it was assumed that the tertiary effluent will be chlorinated for disinfection prior to discharge. Chlorine gas (Cl₂) is used as disinfectant. The chlorine dose was assigned to a uniform distribution of 6-12 mg/L, based on typical chlorine demand for "nitrified activated sludge effluent" (Tchobanoglous et al, 2003). This dose corresponds to effluent coliform concentrations of less than 200 MPN per 100 mL, assuming a contact time of at least 30 min. Chlorination activities consume 27 kWh/d (Menendez, 2010). Dechlorination proceeds via use of sulfur dioxide (SO2), at an assumed dose of 2 mg/L of final effluent (Beal, 2012). Table S11 contains life cycle inventory data for Cl₂, SO₂, and other energy and material inputs.

With respect to TSS and BOD, it was assumed that the concentrations in the secondary effluent pass through the algae pond into the tertiary effluent. This is probably an overly conservative estimate, but there is currently too little data to prove otherwise. It was also necessary to account for the contribution of unsettled algae biomass towards each effluent concentration. For TSS, the algae contribution was accounted for using one minus the capture efficiency during algae thickening (Section 2.3) multiplied by the total algae yield and divided by the total volume of tertiary effluent. This concentration was then converted into a BOD concentration based on the assumed algae stoichiometry and Equation S3.

Equation S3	For $C_a H_b O_c N_d$
	n = moles of O ₂ demanded by 1 mole of C _a H _b O _c N _d
	n = a + 0.25b - 0.5c + 1.25d

For the algae stoichiometry used in this study ($C_{106}H_{181}O_{45}N_{15}$), 148 moles of O_2 are demanded per mole of unsettled algae biomass. Multiplying this ratio by the concentration of algae in the tertiary effluent and by the molecular weight of O_2 and one minus the ash content of algae, the algae TSS concentration is converted into an equivalent algae BOD concentration. For both TSS and BOD, the algae contribution to the tertiary effluent concentration was added together with the pass-through component from the secondary effluent.

For TN and TP, it was assumed that algae growth would consume a significant fraction of the TN and TP entering the pond via the secondary effluent (see Section 2.3). The residual nutrient concentrations were assumed to pass through the pond. For TN, the pass-through nitrogen was added to the nitrogen content of the algae TSS concentration in the tertiary effluent. The algae nitrogen content was roughly 9%, based on the algae stoichiometry assumed in this study. These two concentrations combined were, on average, almost exactly equal to the permitted TN concentration in Table S1; such that partial denitrification was required for roughly 50% of the Monte Carlo simulations. For TP, the pass-through phosphorus was added to the phosphorus concentration of the algae TSS concentration in the tertiary effluent. The phosphorus content was roughly 1% of the algae biomass, based on the stoichiometry assumed in this study. These two concentration in Table S1; therefore, it was assumed that alum is added to bring the TP down to the permitted level.

Finally, with respect to the concentrations of E1, E2, and EE2, these chemicals are not currently regulated in WWTP discharges. Therefore, no attempt was made to reduce their concentrations below what flows out of the algae cultivation pond. These concentrations were computed based on typical

concentrations in municipal secondary effluent and the expected removal of each compound during algae-based treatment. Section 4 contains more information on these calculations.

3 Analysis of Conventional Tertiary Treatment Methods

3.1 Model Overview

Four conventional tertiary treatments were analyzed using LCA, to provide benchmarks for the results from the proposed WWTP + algae system. The evaluated conventional treatments include: ozonation (OZ), ultraviolet irradiation (UV), and adsorption onto granulated activated carbon (GAC). These analyses had the same FU as was used applied to the integrated WWTP + algae system, to ensure meaningful comparison between the system of interest and its selected benchmarks.

3.2 Ozonation (OZ)

Ozonation is widely used as a means of microbial disinfection; however, it also offers some removal of unregulated emerging contaminants. The principal design parameter for most OZ systems is the desired reduction of selected microbial organisms. Based on the assumed fecal coliform concentrations in the secondary effluent (Table S2) and the desired tertiary effluent (200 MPN/100 mL), the ozone dose required for disinfection was assigned to a uniform distribution over the range 4-10 mg/L (Tchobanoglous et al, 2003). This is a typical dose for "activated sludge effluent" given the assumed influent and effluent coliform concentrations. It was also assumed that the removal of estrogenic compounds would exert additional ozone demand. This demand was assigned to a uniform distribution over the range 2.5-3 mg/L, based on previously published studies of ozone-mediated estrogen removal (Maniero et al 2008; Nakada et al, 2007). The transfer efficiency of the ozone gas into the liquid was assigned to a uniform distribution over the range 0.8-0.9 (Tchobanoglous et al, 2003). The total amount of ozone required per functional unit was computed by multiplying the total ozone dose (for coliforms + estrogens) by the total flow rate of secondary effluent and dividing by the gas transfer efficiency.

Consistent with typical practice, it was assumed that ozone would be produced onsite from ambient air. Four steps are required: air preparation, ozone generation, ozone contacting, and residual ozone destruction. Table S6 summarizes the input distributions for the electricity consumption corresponding to each of these four steps. It was assumed that the HRT for ozonation would be roughly 1-30 min, but this was not explicitly accounted for in the modeling calculations.

Table S6. Ozonation electricity	demands,	by stage. ^a
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Stage	Value	Source
Air preparation (kWh/g ozone)	U (4.4, 6.6)	Tchobanoglous et al, 2003
Ozone generation (kWh/g ozone)	U (13.2, 19.8)	Tchobanoglous et al, 2003
Ozone contacting (kWh/g ozone)	U (2.2, 6.6)	Tchobanoglous et al, 2003
Ozone destruction and other (kWh/g ozone)	U (1.2, 2.2)	Tchobanoglous et al, 2003

^a Uniform distributions are denoted "U (min, max)".

3.3 UV Irradiation (UV)

Similar to ozonation, ultraviolet (UV) irradiation is widely used as a means of microbial disinfection and also offers some removal of unregulated emerging contaminants. Lamps emitting the so-called "germicidal fraction" of the UV range, 220-320 nm ("UV-C"), are especially usefully for disinfection purposes. Addition of hydrogen peroxide (H_2O_2) increases the efficiency of microbial disinfection and can also assist with removal of certain low-concentration organic contaminants (e.g., estrogens), by increasing the production of hydroxide radicals (OH•) (Pereira et al, 2007).

As with OZ, the principal design parameter for the UV system modeled in this study was the desired reduction of selected microbial organisms. Based on the assumed concentrations of fecal coliform bacteria in the secondary effluent (Table S2) and the desired tertiary effluent (200 MPN/100 mL), log removals on the order of 2.2 to 4.4 are required. These removals were used to select UV doses suitable for "unfiltered secondary wastewater" with assumed TSS concentrations of approximately 20 mg/L (Darby et al, 1993; Tchobanoglous et al, 2003). The resulting UV doses were assigned to a uniform distribution over the range 25-37 mJ/cm².

From Equation S4, UV dose (D) is the product of lamp intensity (I) times UV contact time (t).

Equation S4 $D = I \times t$

From Equation S5, intensity is a function of lamp characteristics and distance from the irradiated surface.

Equation S5
$$I = I_0 \times \frac{(1 - e^{-kd})}{kd}$$

 I_0 is the intensity of the lamp output at 254 nm directly at the lamp surface, *d* is the depth of the sample away from the lamp surface, and k is the absorbance coefficient. For this study, it was assumed that typical lamp diameters are uniformly distributed over the range 15-20 mm and that the center-to-center distance between adjacent lamps is 75 mm. For this configuration, the maximum value of *d* is roughly 2.6 cm. For nitrified secondary effluents, a typical *k* is 2.303 × a value on the uniform range 0.15-0.35. Once multiplied together, the resulting k has units of 1/cm (Tchobanoglous et al, 2003). Finally, I₀ was computed using "typical" lamp geometries and attributes corresponding to low-pressure, low-intensity mercury lamps: I₀ is triangularly distributed, with min = 25 W, likeliest = 26 W, and max = 27 W; lamp length is uniformly distributed over 1.5-1.8 m (White et al, 1986; Tchobanoglous et al, 2003).

Dividing the lamp's output by its surface area (as defined using length and diameter), converts I_0 from units of W to W/cm². Plugging this value in Equation S5 gives the required lamp intensity (I), also in units of W/m². This value can then be substituted into Equation S4, with the D value specified above, to determine what contact time is required during UV treatment. For the lamps modeled in this study, which was largely based on White et al (1986), the required contact times are roughly several seconds.

The corresponding hydraulic loading rate for the lamps described in the previous paragraph was assigned to a triangular distribution: min = 33, likeliest = 43, and max = 54 L/min-lamp (White et al, 1986). Dividing this value into the daily flow rate corresponding to the FU yields the number of lamps required. It was assumed that these lamps have a nominal power rating given by the following triangular distribution: min = 70 W, likeliest = 80 W, and max = 100 W (White et al, 1986; Tchobanoglous et al, 2003). It was assumed that the lamps operate continuously, such that amount of electricity consumed by the lamps was computing by multiplying their power rating times the number of seconds per year.

Initial H_2O_2 dosing was assigned to the uniform distribution 5-15 mg/L, based on typical installations (Ratnayaka et al, 2009). Estrogen removal reactions under UV + H_2O_2 treatment were modeled according to the pseudo-first order rate model given by Equation S6 (Rosenfeldt and Linden, 2004; Pereira et al, 2007; Crittenden et al, 2012).

Equation S6
$$C_t = C_o \times e^{-k_1 \times t}$$

 C_0 and C_t are concentrations of the target estrogen at times 0 and t, respectively. k_1 is the initial pseudofirst order rate constant for indirect photolysis of the estrogen during reactions mediated by UV + H₂O₂. This parameter has units of inverse time. It is computed according to Equation S7.

Equation S7 $k_1 = k_{1-OH} \times [OH_{SS}^{\bullet}]$

The magnitude of k_1 is given by the product of a second-order rate constant (k_{1-OH}) and the steady-state concentration of hydroxyl radicals (OH*_{SS}). The second-order rate constants were taken from previously published literature for the estrogen targets of interest and assigned to triangular distributions. For E2, k_{1-OH} was triangularly distributed with min = 1.1E10 L/mol-s, likeliest = 1.4E10 L/mol-s, and max = 1.7E10 L/mol-s (Rosenfeldt and Linden, 2004). For EE2, k_{1-OH} was triangularly distributed with min = 8.5E9 L/mol-s, likeliest = 1.1E10 L/mol-s, and max = 1.3E10 L/mol-s (Huber et al, 2003; Rosenfeldt and Linden, 2004). Finally, for E1, k_{1-OH} was triangularly distributed with min = 1.1E10 L/mol-s, likeliest = 2.6E10 L/mol-s, and max = 7.0E10 L/mol-s (Nakonechny et al, 2008). The steady-state concentration of hydroxyl radicals was computed using Equation S8 (Crittenden et al, 2012).

Equation S8

$$[OH_{SS}^{\bullet}] = \frac{2 \times \phi_{H_2O_2} E_0 f_{H_2O_2}}{k_{H_2O_2} [H_2O_2] + k_{HCO_3^-} [HCO_3^-] + k_{DOC} [DOC] + \sum k_{1-OH} \times [Estrogen]}$$

 ϕ_{H2O2} is the quantum yield for hydrogen peroxide, with a value of 0.5 mol/einstein (Crittenden et al, 2012). E_0 is the UV light intensity, in units of einstein/L-s. This parameter is computed using Equation S9.

$$E^0 = \frac{I_0}{N_A V h v}$$

 I_0 is the UV intensity per lamp, as defined for Equation S5, in units of W. N_A is Avogadro's constant (6.023E23 photons/mol). V is the hydraulic flow rate per lamp, as defined above. h is Plank's constant, 6.62E-34 J-s. v is the frequency of light corresponding to 254 nm = 1.18E15 s⁻¹.

 f_{H2O2} is the fraction of light absorbed by H₂O₂, as given by Equation S10. This calculation assumes that all light is absorbed by the water matrix instead of the walls of the reactor and accounts for competing light absorption by dissolved organic carbon (DOC).

Equation S10
$$f_{H_2O_2} = \frac{\varepsilon_{H_2O_2}[H_2O_2]}{\varepsilon_{H_2O_2}[H_2O_2] + \varepsilon_{DOC}[DOC]}$$

 ε_{H2O2} is the molar extinction coefficient for hydrogen peroxide, 19.6 L/mol-cm (Crittenden et al, 2012). [H_2O_2] is the initial concentration of hydrogen peroxide in M units. ε_{DOC} is the molar extinction coefficient for DOC, 0.02 L/mg-cm (Crittenden et al, 2012). [DOC] is the concentration of dissolved organic carbon in units of mg/L. This quantity was estimated based on BOD concentration in the secondary effluent (Table S10), as multiplied by an assumed ratio of BOD to DOC. This ratio was assigned to a uniform distribution over the range 0.2-0.5 (Tchobanoglous et al, 2003).

The denominator of Equation S8 accounts for all reactions that consume hydroxyl radicals by summing together the products of second-order rate constants times their respective initial concentrations. Values for the rate constants were taken from literature: $k_{H2O2} = 2.7E7$ L/mol-s (Crittenden et al, 2012); $k_{HCO3} = 8.5E6$ L/mol-s (Crittenden et al, 2012); $k_{DOC} = 3.9E8$ L/mol C-s (Crittenden et al, 2012); and k_{1-OH} values for E2, EE2, and E1 are assigned to the same values as in Equation S7. $[H_2O_2]$ is converted to M units from the value referenced above. $[HCO_3^-]$ concentration was computed based on the assumed alkalinity of the secondary effluent, 50-100 mg/L as CaCO₃ (Table S10). For this calculation, it was assumed that the pH of the secondary effluent is on the range 6.4-7.2 such that all of the carbonate alkalinity exists as HCO₃⁻. [DOC] was the same as for Equation S10, as computed from the effluent BOD concentration. Finally, concentrations of individual estrogens (E2, EE2, E1) are noted in Table S2. Following calculation of $[OH^{\bullet}_{SS}]$, this value was used to compute pseudo-first order rate constants for each estrogen, according to Equation S7. Average computed values for these parameters (k_1) were 0.017 1/s for E2, 0.013 1/s for EE2, and 0.043 1/s for E1. These values were then used in conjunction with Equation S6 and the estimated UV retention time to estimate estrogen concentrations in the tertiary effluent.

3.4 Adsorption onto Granular Activated Carbon (GAC)

Activated carbon is used extensively as an adsorption material for various wastewater contaminants, including steroid hormones and pharmaceuticals. A downflow fixed-bed granular activated carbon (GAC) reactor was chosen for this study, because this is one of the most common types of adsorption columns for wastewater treatment' by virtue of several key operational advantages compared to other configurations (Sundstrom and Klei, 1979). Additionally, GAC was used instead of powdered activated carbon because it exhibits better removal efficiency for wastewater contaminants and has higher contact surface area (Tchobanoglous, 2003).

It was decided that the daily flow rate corresponding to the FU for this study should be allocated over 14 parallel GAC columns, because this value yields a per-column flow rate (roughly 2,700 m³/d) in the desirable range of hydraulic loadings (1,200-9,600 m³/d) for this reactor configuration (Tchobanoglous et al, 2003). It was also assumed that the residence time in the reactor (based on empty bed contact time – "EBCD") should be uniformly distributed over the range 5-30 min (Tchobanoglous et al, 2003). The cross-sectional area of the mass transfer zone (MTZ) was likewise assigned to a uniform distribution over the range 5-30 m² (Tchobanoglous et al, 2003). With assignment of these parameters, it was then possible to compute the volume per GAC column, the height of the MTZ, and the linear approach velocity of the liquid through the MTZ. These values were compared with typical ranges as reported in Tchobanoglous et al (2003).

Once the dimensions of the MTZ had been computed, it was then possible to compute how much GAC packing is required per FU. The density of the packing was assigned to a uniform distribution over the range 350-550 kg/m³ (Tchobanoglous et al, 2003). The useful life of the packing material was assigned to a triangular distribution: min = 0.3, likeliest = 1.5, and max = 1.7 years (Tchobanoglous et al, 2003). Multiplying the number of columns required per FU by the dimensions of each column and the density of packing, and then dividing by the packing's useful life, yields the amount of packing required to treat the FU flow rate over one year.

Electricity consumption for the GAC tertiary treatment was computed using the Rose Formulation for head loss (h) through a porous medium, as summarized in Equation S11. Table S7 summarizes the parameters required for this calculation.

$$h = \frac{1.067}{\phi} C_d \frac{l}{\alpha^4} \frac{L}{d} \frac{v_s^2}{g}$$

Where:

$$C_d = \frac{24}{N_R} + \frac{3}{\sqrt{N_R}} + 0.34$$
 and $N_R = \phi \frac{dv_s}{v}$

Table S7. Parameters re	quired to com	pute electricity of	consumption durin	g GAC treatment. ^a
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Parameter	Value	Notes
φ , Particle shape factor	0.73	For "crushed coal".
α , Porosity	U (0.38, 0.42)	
L, height of the MTZ (m)	NA	Computed, see preceding text.
d, Grain size diameter (m)	U (0.0008, 0.0009)	
v _s , Approach velocity (m/s)	NA	Computed, see preceding text.
g, Acceleration of gravity (m/s^2)	9.81	
v, Kinematic viscosity of water (m ² /s)	1.003 × 10 ⁻⁶	At 20 °C.

^a Uniform distributions are denoted "U (min, max)". All data are from Tchobanoglous et al (2003).

The head loss computed using Equation S11, in conjunction with the parameters from Table S7, was roughly 1 m. This was converted to pump electricity consumption based on the flow rate per column

and an assumed pump efficiency of 85%. Multiplying this through by the total of number of columns required yields the electricity consumed for GAC treatment of the FU over one year. Additionally, it was assumed that the columns must be backwashed and air scoured monthly; however, this was not explicitly accounted for in this analysis.

3.5 Anaerobic Digestion and Tertiary Effluent Quality for the Conventional Treatments

3.6.1 Anaerobic Digestion

As in the system incorporating algae-mediated tertiary treatment, it was assumed that the primary and secondary solids from the systems incorporating conventional tertiary treatments would be anaerobically digested to produce methane-derived electricity. The digestion parameters corresponding to primary and secondary solids in Table S4 were used to compute how much electricity could be produced via anaerobic digestion, what amounts of heat and electricity are required to operate the digester and the belt filter press.

3.6.2 Effluent Quality – Microbial

For all three conventional treatments (OZ, UV, GAC), it was assumed that the flow rate of secondary effluent is exactly equal to the flow rate of discharged tertiary effluent. Because the OZ and UV treatments are, by design, disinfection treatments, no additional treatment was required to bring the fecal coliform concentration down to the permitted level in Table S1. In contrast, it was assumed that the GAC effluent would require chlorination using the same disinfectant (Cl₂) as was used in the algae-based system. The chlorine dose required to achieve 200 MPN/100 mL in "filtered nitrified effluent" is uniformly distributed over the range 6-12 mg/L, assuming a 30-min contact time (Tchobanoglous et al, 2003). Dechlorination is achieved using SO₂. Table S11 contains life cycle inventory data for Cl₂, SO₂, and other energy and material inputs.

3.6.3 Effluent Quality – TSS, BOD, TN, and TP

TSS, BOD, TN, and TP removal efficiencies for all three conventional treatments (OZ, UV, GAC) were collected from literature sources. These were applied to the concentrations in the secondary effluent to compute what concentrations of each constituent exist in the effluents of each tertiary treatment. Pertinent removal efficiencies are summarized in Table S8.

Treatment	TN Removal (%)	TP Removal (%)	TSS Removal (%)	BOD Removal (%)	Sources
OZ	Tri (2, 14, 20)	0	38	Tri (0, 36, 67)	Paraskeva et al, 2002 Paraskeva et al, 2005 Petala et al, 2006
UV	15	0	0	U (0, 1)	Bems e al, 2008 Lehtola et al, 2003 Paraskeva et al, 2005
GAC	Tri (15, 24, 42)	Tri (72, 88, 94)	21	Tri (34, 81, 90)	

Table S8. Removal efficiencies for TSS, BOD, total nitrogen (TN), and total phosphorus (TP) during the selected tertiary treatments.^a

^a Triangular distributions are denoted "Tri (min, likeliest, max)". Uniform distributions are denoted "U (min, max)".

4 Estrogenicity Calculations

The representative and widely ubiquitous estrogens were included in this study: 17β -estradiol, 17α -ethinylestradiol, and estrone. Figure S2 presents chemical structures for these compounds.



Figure S2. Chemical structures for the estrogenic steroid hormones included in this study.

Estrogenicity removal in each system was computed based on the assumed distributions of influent estrogen concentrations (Table S2), individual estrogen removal efficiencies from previously published studies of the modeled tertiary treatments (Figures S3 and S4 and Table S9), and estrogenic toxicities of the three selected estrogen species (Table S10). The latter were reported in units of "nanograms estradiol equivalent per liter", ng EEQ/L. The residual concentrations of each selected estrogen (Table S9) were multiplied by their relative estrogenic potencies (Table S10), and these quantities were summed together to compute overall estrogenic toxicity for the WWTP influent and effluent. Decreases in the concentration of E2, E1, or EE2 from influent to effluent are thus associated with a corresponding decrease in estrogenic toxicity.



Figure S3. Literature data for evaluation of algae-mediated estrone (E1) removal kinetics. Data are from Shi et al (2010).



Figure S4. Literature data for evaluation of algae-mediated 17α -ethinylestradiol (EE2) removal kinetics. Data are from: Lai et al (2012), Ge et al (2008), Shi et al (2010), and Della Greca et al (2008).

Table S9. Parameters for determination of residual estrogen concentrations during tertiary treatment:
expected removal efficiencies from previously published studies for algae, OZ, and GAC; and, first-order
rate constants for UV.

Treatment	Estrogen	Removal Efficiency (%)	Pseudo-First Order Rate Constant	Sources
Algae	E2	Tri (64, 70, 80)		Della Greca et al 2008.
Algae	E1	Tri (30, 36, 45)		Ge et al 2008, Lai et al 2002, Shi et al
Algae	EE2	Tri (50, 74.5, 99)		(2010) <i>,</i> This study
OZ	E2	Tri (63, 80, 99)		Filby et al (2010),
OZ	E1	Tri (74, 90l, 98)		Maniero et al (2008),
OZ	EE2	U (64, 99)		Ternes et al (2003)
UV	E2	NA	See Section 4.3	
UV	E1	NA	See Section 4.3	Calculated
UV	EE2	NA	See Section 4.3	
GAC	E2	Tri (39, 64, 90)		
GAC	E1	U (76, 98)		Filby et al (2010), Westerhoff et al (2005)
GAC	EE2	U (53, 57)		

^a Triangular distributions are denoted "Tri (min, likeliest, max)". Uniform distributions are denoted "U (min, max)".

Table S10. Empirical estrogenic potency factors for the three eEDCs evaluated in this study. E2 is assigned an estrogenic potency factor of 1 ng estradiol-equivalent (EEQ) per ng E2 because E2 is the prototypical estrogen and all other eEDC potencies are expressed relative to its potency.

Estrogen species	Estrogenicity (ng EEQ/ng estrogen) ^{a,b}
17β-Estradiol (E2)	L (1.0, 0.001)
Estrone (E1)	L (0.5, 0.05)
17α-Ethinylestradiol (EE2)	L (1.35, 0.135)

^a Data are from Khanal et al (2006), Sumpter and Johnson (2005), Ternes et al (2004), Westerhoff et al (2005).

^b Lognormal distributions are denoted "L (mean, standard deviation).

5 LCA Impact Factors and Supplemental Results

5.1 Database Impact Factors

LCA impact factors from published literature and the industry-standard ecoinvent database (Weidema, 2008) were used to assess materials and energy inputs associated with components of each LCA model. Pertinent LCA impact factors are summarized in Table S11.

Table S11. Life cycle impact factors for materials and energy inputs used in LCA models for the selected tertiary treatment methods (algae, OZ, UV, GAC). Values are shown using μ/σ notation in each column, where μ is mean value and σ is standard deviation. All data were from ecoinvent v2. (Weidema, 2008) unless otherwise noted.

Item	Unit Basis	Energy Use (MJ)
Alum	1 kg Al ₂ (SO ₄) ₃	5.7/1.29
Chlorine gas	1 kg Cl ₂	19.23/NAª
Electricity	1 kWh from US grid	12.5/10.0
Fertilizer - CaH ₂ P ₂ O ₈	1 kg P ₂ O ₅	33.8/14.5
Fertilizer - H ₁₂ N ₃ O ₄ P	1 kg P ₂ O ₅	37.5/5.4
Fertilizer - N ₂ H ₄ CO	1 kg as N	62.1/11.8
Granular Activated Carbon	1 kg from crushed coal	118/17.1
Heating Oil (Light)	1 MJ from light heating oil	1.3/0.2
Hydrogen Peroxide	1 kg as H_2O_2	13.2/NA ^b
Methanol	1 kg CH₃OH	37.5/5.4
Sulfur Dioxide	1 kg SO ₂	5/NA ^c

^a Worrell et al, 2000; ^b AkzoNobel, 2011; ^c Beal et al, 2012.

5.2 Confidence Intervals for Reported Metrics

Table S12. 95% confidence intervals for key environmental metrics for four evaluated tertiary treatment systems: algae cultivation, ozonation (OZ), UV irradiation (UV), and adsorption on granular activated carbon (GAC). Values are from empirical output distributions arising from Monte Carlo sampling. See Table 1 of the paper for mean values.

Metric	WWTP	Algae	OZ	UV	GAC
EROI (MJ energy _{out} /MJ energy _{IN})	[0.26, 0.88]	[0.36, 0.99]	[0.05, 0.63]	[0.11, 0.90]	[0.10, 0.69]
Normalized net energy use – tertiary	NΑ	[-2.6E3,	[6 4F4 9 3F5]	[1.3E5,	[4 6F4 4 8F5]
treatment only (MJ/ g EEQ removed) ^a		1.5E54]		1.1E7]	
Residual estrogenicity (ng EEQ/L) ^b	[13.9, 38.6]	[5.1, 18.6]	[1.7, 7.5]	[12.9, 36.8]	[1.9, 9.2]
Residual BOD concentration (mg/L) ^b	[6.4, 11.1]	[7.2, 13.8]	[3.1, 8.7]	[6.4, 11.1]	[1.1, 5.2]
Residual TSS concentration (mg/L) ^b	[13.1, 16.4]	[13.8, 19.8]	[8.1, 10.2]	[13.1, 16.4]	[10.3, 13.0]

^a <u>Positive</u> net values indicate that more energy is consumed than produced in the system. <u>Negative</u> net values indicate that more energy is produced than consumed.

^b Refers to discharged effluent: secondary for WWTP, tertiary for algae, OZ, UV, and GAC.

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