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Environmental, Human Health, and Economic Implications of Landfill Leachate Treatment for
PFAS Removal

BY

DANYI FENG

B.S. Environmental Engineering, University of New Hampshire, 2018

Master's Thesis

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in

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DEDICATION

I dedicate this work with love to my parents, Hao Feng and Jinhua Wu who have supported me throughout my education. Thanks for making me see this adventure through the end.

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ABSTRACT¹

Landfill leachate is commonly treated offsite with municipal wastewater. This offsite leachate treatment may be limited or no longer applicable due to the increasingly stringent regulations and concerns related to PFAS discharge into the environment, resulting in development of full-scale, onsite leachate treatment facilities. To help landfills prepare for the potential shift from offsite to onsite leachate treatment for PFAS compliance, this study analyzed and compared the environmental, human health, and economic performances of a typical onsite and a typical offsite leachate treatment alternative using life cycle assessment (LCA) and life cycle cost assessment (LCCA). Two distinct functional units were investigated: 1 m³ of leachate treated and 1 g of PFAS removed. Using a landfill site located in Zhuzhou, China as a testbed, we tested two hypotheses: 1) environmental, human health, and economic tradeoffs exist between onsite and offsite treatment scenarios; 2) the tradeoffs change when different functional units are used. Our results show that the onsite scenario offers benefits from human health and economic perspectives, while the offsite scenario performs general better environmental outcomes. The extent of this tradeoff varies when different functional units were adopted and varies depending on PFAS concentrations in raw leachate.

¹ The work presented in this MS thesis should be cited as the following manuscript in preparation: Danyi Feng, Cuihong Song, and Weiwei Mo, Environmental, Human Health, and Economic Implications of Landfill Leachate Treatment for PFAS Removal, manuscript in preparation.

Chapter 1. Introduction

Every year, a large amount of leachate is produced in landfills worldwide as rainwater infiltrates through solid wastes (Amaral et al., 2016; Sadri et al., 2008). In the US alone, nearly 61 million m³ of leachate were generated in 2013 (Lang et al., 2017). The volume of leachate generation is anticipated to increase as the amount of landfilled solid waste continues to grow globally (Deng and Englehardt, 2006; Hoornweg and Bhada-Tata, 2012). Leachate is a highly contaminated and toxic liquid containing a variety of pollutants such as dissolved organic matters (e.g., chemical oxygen demand (COD), organic nitrogen), heavy metals (e.g., Zn, Cu, Pb), xenobiotic organic compounds (e.g., phenols), and inorganic salts (e.g., Na⁺, NH₄⁺) (Aziz et al., 2010; Baun et al., 2004; Kjeldsen et al., 2002; Ogata et al., 2016). The composition and concentration of landfill leachate can vary significantly depending on waste composition and moisture content, landfill age and type, and meteorological condition. Landfill leachate has also been reported as a major source of per- and polyfluoroalkyl substances (PFAS) (Masoner et al., 2020). PFAS is a group of emerging contaminants that has attracted attention worldwide given their ubiquitous distribution, environmental persistence, and severe ecosystem and human health impacts (e.g., cancer, immune system weakening, and thyroid hormone disruption) (Grandjean and Clapp, 2015; Wei et al., 2019). Previously reported PFAS concentrations in landfill leachate ranged from 5.1 to 298,559 ng/L across the world (Wei et al., 2019).

Landfill leachate is commonly transported to wastewater treatment plants (WWTPs) and treated together with municipal wastewater (hereafter referred to as offsite treatment) (Bilardi et al. 2018). The offsite treatment method currently applies to around 62% of the leachate generated in the US (Dereli et al., 2020). This percentage goes up to 100% in the Republic of Ireland

(McCarthy et al., 2010; Zhao et al., 2018). Nevertheless, WWTPs that employ conventional treatment technologies (e.g., biological treatment, oxidation, and coagulation-flocculation) are generally ineffective in PFAS removal (Wei et al., 2019). Chen et al. (2018) reported PFAS removal efficiencies in WWTPs to range from 1.5-32.2%. Gallen et al. (2018), on the other hand, reported elevated PFAS concentrations in the WWTP effluent as compared to influent. This is likely a result of the degradation of precursor chemicals during the WWTP's biological treatment process.

Meanwhile, regulations of PFAS compounds are rapidly evolving over the past few years. In 2016, the US Environmental Protection Agency (EPA) established drinking water Lifetime Health Advisory Levels for two groups of PFAS compounds, perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) at 70 ng/L, separately or combined (CRS 2019). Since then, many states (e.g., California, New Jersey, Vermont, and New Hampshire) have issued more stringent drinking water PFAS Maximum Contaminant Levels (MCLs) that cover a broader number of PFAS compounds (AWWA 2020a). For instance, New Hampshire established MCLs of 12 ng/L for PFOA, 15 ng/L for PFOS, 18 ng/L for perfluorohexane sulfonic acid (PFHxS), and 11 ng/L for perfluorononanoic acid (PFNA). These drinking water MCLs also simultaneously update the state's ambient groundwater quality standards, which apply to wastewater effluent discharges to groundwater. Additionally, three states including Michigan, Montana, and Oregon have regulated PFAS discharges into surface waters for protection of drinking water sources (AWWA 2020b). These PFAS regulations in ambient ground and surface water bodies imply the potential need of actions be taken by the WWTPs and landfills for enhanced PFAS removal or treatment. For those WWTPs that accept landfill leachate with high

PFAS concentrations, they may no longer be able to do so under these regulations. In response, landfills may need to develop onsite leachate treatment facilities. Onsite treatment systems refer to full-scale treatment systems constructed onsite of landfills (Townsend et al., 2015). They adopt a wide range of treatment technologies, including total recirculation, conventional physical and chemical treatment, or multi-stage membrane treatment that is capable of providing near-complete removal of contaminants (Calabrò et al., 2018; Renou et al., 2008; Robinson, 2007; Zhao et al., 2012). The near-complete removal facilities allow effluent to be directly discharged into the environment, and hence are the most suitable for addressing the need of PFAS treatment/removal. However, such treatment facilities often require substantial constructional and operational investments (Zhang et al., 2019), which can impose a significant financial burden on landfill owners.

To help stakeholders prepare for the potential transition, an enhanced understanding of environmental, human health, and economic comparisons of offsite and onsite leachate treatment scenarios considering PFAS removal is imperative. Life cycle assessment (LCA) and life cycle cost assessment (LCCA) are widely-used tools to quantify the life-cycle environmental and economic impacts of a product or system throughout raw material extraction, equipment manufacturing, use, and disposal (Liamsanguan and Gheewala, 2008). The application of LCA and LCCA in landfill leachate treatment systems is still very limited. To our knowledge, Di Maria and Sisani (2017) and Di Maria et al. (2018) are the only two studies that compared the environmental impacts of onsite (33% treated onsite and 67% treated offsite) and offsite (100% treated offsite) leachate treatment scenarios. Three different onsite leachate treatment schemes were considered including evaporation, RO pre-treatment plus evaporation, and 3-stage RO

treatment systems, while offsite treatment was primarily a traditional activated sludge process. The 3-stage RO treatment system combined with offsite treatment was found to perform the best in all environmental impact categories. Very few studies investigated both the environmental and economic performances of onsite leachate treatment systems. Postacchini et al. (2018) assessed an onsite leachate treatment system that includes coagulation, flocculation, activated carbon filtration, and ion exchange, and highlighted potential environmental and economic tradeoffs as a result of treatment chemical selections. Most existing leachate treatment LCA studies used either 1 m³ of raw leachate treated (Di Maria et al., 2018; Di Maria and Sisani, 2017; Postacchini et al., 2018) or one average person in a reference year (Xing et al., 2013) as a functional unit (FU). A few LCA studies of wastewater treatment systems compared outcomes in terms of both volume of wastewater treated and mass of contaminants/nutrients removed (Delre et al., 2019; Rashid et al., 2020; Real et al., 2017; Rodriguez-Garcia et al., 2011). They found that using the different FUs may result in significantly different environmental, human health, and economic tradeoffs, highlighting the importance of balancing various decision objectives and creating win-win decision solutions. However, none of these studies have utilized the mass of PFAS treated as a FU.

Accordingly, this research aims to quantify the potential environmental, human health, and economic implications when landfills shift from offsite to onsite leachate treatment for PFAS compliance. To achieve this, we analyzed and compared the environmental, human health, and economic performances of a typical onsite and a typical offsite leachate treatment alternative using LCA and LCCA using a landfill site located in Zhuzhou, China as a testbed. Two distinct functional units were investigated and compared: 1 m³ of leachate treated and 1 g of PFAS

treated. Two hypotheses were tested: 1) environmental, human health, and economic tradeoffs exist between onsite and offsite treatment scenarios; 2) the tradeoffs change when different functional units are used.

Chapter 2. Method

The following sections provide a brief background of the landfill site (Section 2.1), a detailed description of the two leachate treatment scenarios (Section 2.2), methods utilized for life cycle environmental and health impact assessment (Section 2.3) and economic assessment (Section 2.4), and methods used for sensitivity analysis (Section 2.5).

2.1. Study site description

Nanjiao landfill, located in the City of Zhuzhou, China, was selected as a testbed system given the detailed treatment and material and energy usage data are available to the authors. The landfill served a population of 4 million (HP, 2020) with an area of 93,500 m². It can support 3.6 million metric tons of wastes with an originally planned service life of 16.7 years. Since its opening in 2003, around 1,100 metric tons of wastes were dumped into this site every day, which led to an early saturation in 2014. The landfill was closed in 2018 and was capped and lined after closure. The climate in Zhuzhou is warm and temperate with a significant amount of rainfall throughout the year. The annual average temperature is around 16 °C to 18 °C and the average annual cumulative rainfall is approximately 1,500 mm (CMA, 2020). In 2018, Nanjiao landfill generated an average of 300 m³ of leachate per day. The average measured levels of COD, BOD₅, total suspended solids (TSS), total nitrogen (TN), ammonia nitrogen, and total phosphorus (TP) in the leachate were around 1,500, 340, 700, 671, 495, and 3 mg/L,

respectively. Because PFAS is not monitored or measured by Nanjiao landfill, we assumed the PFAS concentration in raw leachate to be 150,704 ng/L, which is an average value of the previously reported PFAS concentration range (2,849 to 298,559 ng/L) in China's landfills (Wei et al., 2019).

2.2. Description of the two leachate treatment scenarios

The onsite scenario is currently adopted by Nanjiao landfill. This leachate treatment system consists of neutralization, coagulation-flocculation, up-flow anaerobic sludge blanket filter (UASB), 2-stage Anaerobic and Aerobic (A/O) reactors, and an additional 3-stage membrane treatment process that includes a membrane bioreactor (MBR) system, nanofiltration (NF), and reverse osmosis (RO) (Figure 1). The MBR system has built-in ultrafiltration membrane in two 2.7 m × 3 m × 6.5 m reactors. Nine types of chemicals are used in this scenario for system operation and maintenance, including aluminum sulfate ($\text{Al}_2(\text{SO}_4)_3$), polyacrylamide (PAM), hydrochloric acid (HCl), sodium hydroxide (NaOH), glucose, organosilicon polymer, amino trimethylene phosphonic acid (ATMP), sodium hypochlorite (NaClO), and citric acid. Specific uses of these chemicals are provided in Table 1. The landfill reported removal efficiencies of the main effluent quality parameters (e.g., COD, TN, and TP) to be around 99% (BETC 2019). We assumed the same removal efficiency for PFAS, given RO has been previously reported to be able to remove 99% of PFOA and PFOS (Flores et al., 2013). The treated leachate is directly discharged into the Xiang River, and the retentate is recirculated back to the landfill. Sludge is disposed in the landfill without any treatment (e.g., thickening and dewatering).

The offsite scenario is a hypothetical scenario consisting of leachate onsite storage, truck transportation, and leachate treatment in a local WWTP (Figure 1). The onsite storage tank was assumed to be 400 m³ to provide a sufficient buffer for storing the volume of leachate generated in one day. The 12-ton vacuum truck was assumed to transport leachate daily to the Longquan WWTP located 9.5 km away from Nanjiao landfill. The size of the truck was selected based on the maximum truck volume allowed to pass the lowest height of bridges and tunnels in the city. This WWTP has an average flow rate of 100,000 m³/day. Its treatment processes consist of pretreatment (fine screen and grit chamber), 3-stage Anaerobic/Anoxic/Aerobic (A/A/O) treatment, MBR, and UV disinfection. Five types of chemicals are used in the treatment processes, including Al₂(SO₄)₃, ferrous sulfate (FeSO₄), NaOH, critic acid, and NaClO (specific chemical uses provided in Table 1). The Longquan WWTP has relatively high COD, BOD₅, TSS, TP, and TN removal efficiencies at 92.3%, 99%, 97.3%, 86.2% and 62.3%, respectively, according to the plant's annual report in 2018. Heavy metals were not detected in either the influent or the effluent. The Longquan WWTP does not monitor for its PFAS concentration or removal efficiency. A previous study reported a 21% PFAS removal efficiency for a WWTP that has the identical treatment process as the Longquan WWTP (Pan et al., 2016). Effluent of the Longquan WWTP is also discharged into the Xiang River.

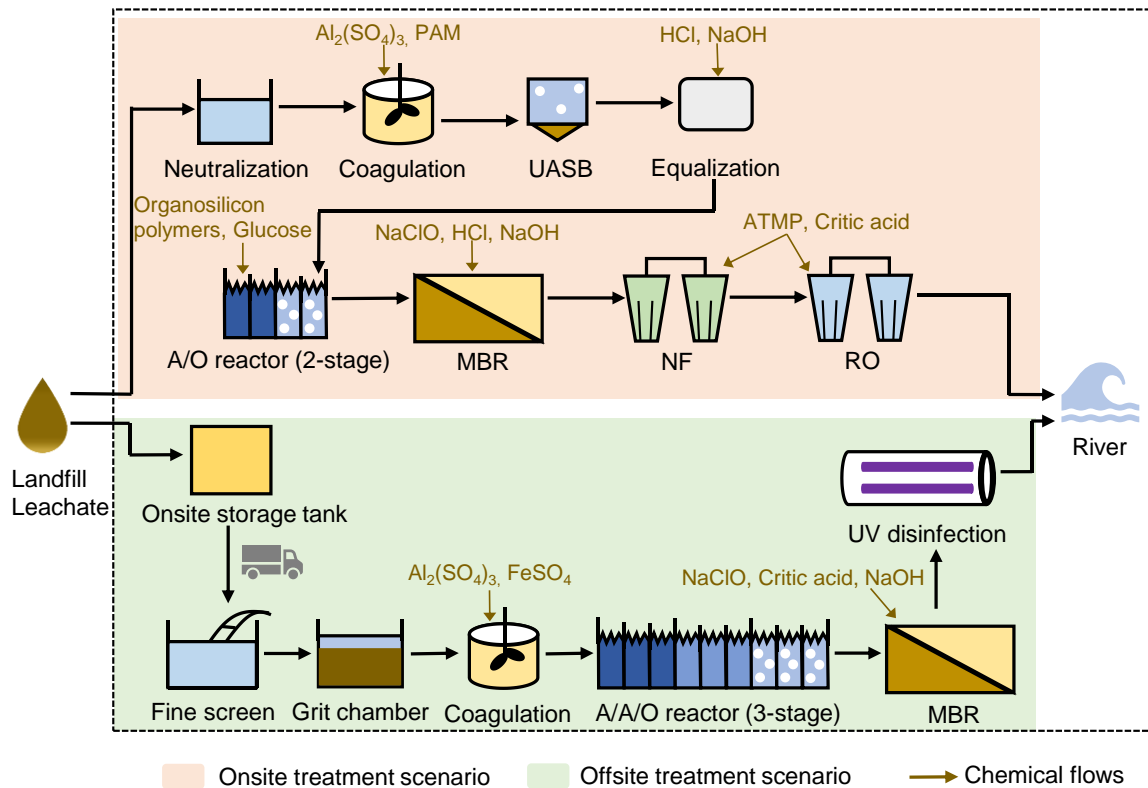


Figure 1. Conceptual process flow diagram for the onsite and offsite leachate treatment scenarios.

2.3. Life cycle environmental and human health assessment

The environmental and human health impacts of the two leachate treatment scenarios were characterized using the Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts (TRACI) version 2.1. TRACI is a mid-point life cycle impact assessment method developed by the US EPA (Ryberg et al., 2014). It provides characterization factors to quantify seven environmental indicators (ozone depletion (OD), global warming (GW), smog (SM), acidification (AC), eutrophication (EU), ecotoxicity (ET), fossil fuel depletion (FFD)), and three human health indicators (respiratory effects (RE), human health-carcinogens (HHc), and human health-noncarcinogens (HHnc)). The analysis was conducted primarily through SimaPro 9.0, supplemented by the Economic Input-Output Life Cycle Assessment (EIO-LCA) web tool (CMU 2018). The assessment adopted a 15-year time horizon as this is a typically assumed

lifespan for key fixed assets in this study (Pirsaheb et al., 2016). A system boundary of construction, operation, and maintenance phases was considered. It has to be noted that because the Longquan WWTP was already in place, its construction was excluded from analysis for the offsite scenario. The end-of-life phase was neglected for both scenarios because the treatment plants are usually refurbished onsite and not fully dismantled (Postacchini et al., 2018).

The characterized result of a certain environmental or human health impact (I_{LC}) was calculated and scaled to one FU using Equation 1.

$$I_{LC} = \frac{I_c + \sum_{t=1}^{15}(I_{o,t} + I_{m,t})}{K_{LC}} \quad \text{Equation 1}$$

where, I_c represents the total impact of the construction phase. $I_{o,t}$ and $I_{m,t}$ are the total impacts of the operation phase and the maintenance phase in year t , respectively. K_{LC} is either the total volume of raw leachate being treated in a 15-year lifespan which is around 1,650,000 m³, or the total amount of PFAS treated in a 15-year lifespan which equals 246,175 g for the onsite scenario and 52,219 g for the offsite scenario. The total amount of PFAS being treated was calculated as the product of PFAS concentration in raw leachate, total volume of raw leachate being treated, and removal efficiencies of PFAS under the two scenarios. Calculations of I_c , $I_{o,t}$ and $I_{m,t}$ for the onsite and the offsite scenarios were detailed in the following subsections.

2.3.1. Onsite treatment scenario

A detailed inventory of the onsite scenario was provided in Table 1. The construction impact of the onsite scenario was calculated using the EIO-LCA web tool (CMU 2018) and the construction cost was obtained from Nanjiao landfill (Nanjiao Landfill 2018). Particularly, the “other nonresidential structure” sector in the US 2002 Producer Price Model was used for this

analysis. The 2002 model was selected because it is the latest model that has incorporated the TRACI method. Cost data were obtained in 2018 Chinese Yuan (RMB). They were first converted to US dollars (USD) using the 2018 RMB/USD currency rate (6.8 RMB/USD) (SAFE 2018), and then converted to 2002 US dollar values using the Consumer Price Index obtained from US Bureau of Labor Statistics (2020) (refer Table S1 in the Appendix for details). The TRACI method embedded in SimaPro and EIO-LCA web tool uses different units for HHc (CTUh vs. kg benzene eq), HHnc (CTUh vs. kg toluene eq), RE (PM10 eq vs. PM2.5 eq), and ET (CTUe vs. kg 2,4D). We converted the outcomes of these four impact categories to align with the units being reported through SimaPro. The conversion factors used were 0.23 PM10 eq/PM2.5 eq for RE, 2.42×10^{-7} CTUh/kg benzene eq for HHc, 1.78×10^{-8} CTUh/kg toluene eq for HHnc, and 860 CTUe/kg 2,4D for ET (Thiel et al. 2015). Additionally, averaged values of the high and low outputs obtained for HHc, HHnc, and ET from the EIO-LCA web tool were used in our analysis.

Impacts associated with the operation phase were calculated based on the electricity consumption, chemical usage, and direct water emission data directly obtained from Nanjiao landfill (Table 1). For chemicals and membrane materials not found in SimaPro, their closest resemblances were used. All the direct emissions except for PFAS were found under waterborne emissions in SimaPro, and converted to TRACI impacts. Since there is no impact data related to PFAS compounds in SimaPro, we used polychlorinated biphenyls (PCBs) as a surrogate to estimate the environmental and human health impacts of PFAS compounds, considering that PFAS have been reported to have similar human health effects as PCBs (Petersen et al., 2018). Log K_{ow} (n-octanol/water partition coefficient) is a widely used measure of a chemical's

lipophilicity and hydrophilicity (Sangster, 1997), which also indicates the chemical's bioaccumulation and toxicity characteristics in living organisms and the environment (Cumming and Rücker, 2017). The log K_{ow} values of PFAS have been previously reported to be between 5.3 and 8.4, which is within the range of the log K_{ow} values of PCBs (4.6-9.6) (Eisler and Belisle, 1996; Smith et al., 2016; Zhang et al., 2013). We identified 29 PCB compounds listed under the waterborne emissions in SimaPro (see Table S2 in the Appendix for details). The impacts of each PCB compound were calculated using the TRACI method and the average impact values of the 29 compounds were then used to indicate the impacts of PFAS compounds.

The maintenance phase included chemicals used for membrane cleaning, membrane replacement, and membrane disposal (Table 1). Membrane replacement was assumed to occur every 5 years for the MBR, and every 3 years for NF and RO based on data collected from Nanjiao landfill. New membranes were transported from a manufacturer 840-km away from the onsite treatment plant, and used membranes were assumed to be disposed through landfill.

Table 1. The life cycle inventory of the onsite leachate treatment scenario over a 15-year time horizon.

| Inputs type of emission/resource | Total amount | Unit | SimaPro Entries | Note |
|---|--------------|------------|--|---|
| Construction | | | | |
| Construction | 488,001 | \$2002 USD | EIO-LCA, Construction, other nonresidential structure | |
| Operation | | | | |
| Electricity | 15,973,560 | kWh | Electricity, medium voltage {CN} market group for Alloc Def, S | |
| Sodium Hydroxide (NaOH) | 360,000 | kg | Neutralising agent, sodium hydroxide-equivalent {GLO} market for Alloc Def, S | Adjust pH in equalization pool; clean MBR system |
| Organosilicon polymers | 10,875 | kg | Polystyrene, high impact {GLO} market for Conseq, U | Restraining defoam in biological tank |
| Polyacrylamide (PAM) | 7,500 | kg | Polyacrylamide {GLO} market for Alloc Def, S | Coagulation |
| Hydrogen chloride (HCl) | 30,000 | kg | Hydrochloric acid, without water, in 30% solution state {RER} market for Alloc Def, S | Adjust pH in equalization pool; MBR system cleaning |
| Amino trimethylene phosphonic acid (ATMP) | 18,000 | kg | Phosphoric acid, industrial grade, without water, in 85% solution state {GLO} market for Alloc Def, S | Anti-scaling and descaling in NF & RO operation |

| | | | | |
|---|-----------|------|--|--|
| Glucose | 403,500 | kg | Sugar, from sugarcane {GLO} market for Alloc Def, S | Provide carbon source during anaerobic and aerobic processes |
| Poly aluminium chloride (PAC) | 15,000 | kg | Aluminium sulfate, powder {GLO} market for Alloc Def, S | Coagulation |
| Concentrate Recirculation | 3,079,688 | kWh | Electricity, medium voltage {CN} market group for Alloc Def, S | |
| Maintenance | | | | |
| NaOCl | 15,000 | kg | Sodium hypochlorite, without water, in 15% solution state {GLO} market for Alloc Def, S | Remove fouling from MBR system |
| Citric acid | 30,000 | kg | Citric acid {GLO} market for Alloc Def, S | Clean membranes |
| MBR membrane replacement (PTFE) | 19,440 | kg | Tetrafluoroethylene film, on glass {RER} production Alloc Def, S | |
| NF membrane replacement | 2,465 | kg | Polyamide (Nylon) 6.6/EU-27 | |
| RO membrane replacement | 2,465 | kg | Polyamide (Nylon) 6.6/EU-27 | |
| Transportation for membrane replacement | 20,471 | tkm | Transport, freight, lorry, unspecified {GLO} market for Alloc Def, S | |
| Membrane disposal | 24,370 | kg | Municipal solid waste (waste scenario) {CH} Treatment of municipal solid waste, landfill Alloc Def, S | |
| Direct water emissions | | | | |
| COD | 225 | mg/L | COD, Chemical Oxygen Demand | |
| TSS | 105 | mg/L | Suspended solids, unspecified | |
| TN | 101 | mg/L | Nitrogen, total | |
| TP | 0.45 | mg/L | Phosphorus, total | |
| PFAS | 1,507 | ng/L | Average of 29 PCB compounds | |

2.3.2. Offsite treatment scenario

A detailed life cycle inventory of the offsite treatment scenario was provided in Table 2. Impacts associated with the construction phase was estimated using the EIO-LCA method based on the estimated cost of the onsite leachate storage tank. The operation phase contained electricity consumption, chemical usages, leachate transportation, and direct water emissions. The electricity and chemical usage data to treat 1 m³ of wastewater were obtained from Chen et al. (2018), which studied a WWTP that has a similar treatment capacity and identical treatment processes as the Longquan WWTP. We assumed that adding leachate to the WWTP will not significantly influence its electricity and chemical usages during the treatment process given the leachate flow rate is extremely small compared to the wastewater flow rate (300 m³/day vs. 100,000 m³/day), despite leachate's relatively higher organic strength (1,500 mg/L vs. 157 mg/L of COD). Therefore, the electricity and chemical usages for treating the mixed leachate and

wastewater at the WWTP was assumed to be the same as treating wastewater alone. Effluent water quality was obtained directly from the Longquan WWTP. Direct water emission impacts were estimated using the same process as the onsite scenario. The maintenance phase considered membrane cleaning, replacement (every 5 years), and disposal. Membranes were assumed to be sourced from the same manufacturer 840-km away, and used membranes were disposed through landfill.

Table 2. The life cycle inventory of the offsite scenario over a 15-year time horizon.

| Inputs type of emission/resource | Total amount | Unit | Simapro Entries | Function |
|---|--------------|------------|--|---|
| Construction | | | | |
| Construction | 12,796 | \$2002 USD | EIO-LCA, Construction, other nonresidential structure | |
| Operation | | | | |
| Electricity | 773,850 | kWh | Electricity, medium voltage {CN} market group for Alloc Def, S | |
| Poly aluminium chloride (PAC) | 10,676 | kg | Aluminium sulfate, powder {GLO} market for Alloc Def, S | Coagulant |
| Iron(II) sulfate (FeSO4) | 31,317 | kg | Iron sulfate {GLO} market for Alloc Def, S | Flocculate phosphorus |
| Sodium Hydroxide (NaOH) | 148.5 | kg | Neutralising agent, sodium hydroxide-equivalent {GLO} market for Alloc Def, S | pH adjustment in biological processes; Membrane system cleaning |
| Transportation for leachate transit | 15,603,750 | tkm | Transport, freight, lorry 7.5-16 metric ton, EURO6 {GLO} market for Alloc Def, S | |
| Maintenance | | | | |
| Sodium hypochlorite (NaOCl) | 13,448 | kg | Sodium hypochlorite, without water, in 15% solution state {GLO} market for Alloc Def, S | Remove membrane fouling |
| Citric acid | 808.5 | kg | Citric acid {GLO} market for Alloc Def, S | Clean membranes |
| MBR/PVDF (membrane replacement) | 175 | kg | Polyvinylfluoride, film {RoW} production Alloc Def, S | |
| Transportation for membrane replacement | 147 | tkm | Transport, freight, lorry, unspecified {GLO} market for Alloc Def, S | |
| MBR/PVDF (membrane disposal) | 175 | kg | Municipal solid waste (waste scenario) {CH} Treatment of municipal solid waste, landfill Alloc Def, S | |
| Transportation for membrane disposal | 2 | tkm | Transport, freight, lorry, unspecified {GLO} market for Alloc Def, S | |
| Direct water emissions | | | | |
| COD | 180 | mg/L | COD, Chemical Oxygen Demand | |
| TSS | 36 | mg/L | Suspended solids, unspecified | |
| TN | 138 | mg/L | Nitrogen, total | |
| TP | 4.35 | mg/L | Phosphorus, total | |
| PFAS | 119,056 | ng/L | Average of 29 PCB compounds | |

2.4. Life cycle cost assessment (LCCA)

Life cycle costs of the two treatment scenarios were analyzed using the net present value (NPV) approach using Equation 2 (Lin et al., 2011).

$$NPV_{LC} = \frac{\text{Capital cost} + \sum_{t=1}^{15} \frac{O_t + M_t}{(1+\alpha)^t}}{K_{LC}} \quad \text{Equation 2}$$

where, NPV_{LC} is the life cycle cost for treating 1 m³ of leachate or treating 1 g of PFAS, \$2018 USD/(m³ of treated leachate or g of PFAS removed). As in Equation 1, K_{LC} is the total amount of FUs within the 15-year time horizon. α is the annual discount rate, 6% (Beh et al., 2014; Wu et al., 2015). O_t and M_t are the operation and maintenance costs in year t , respectively. Table 3 provides the capital, operation, and maintenance costs associated with the two scenarios.

Table 3. Capital, operation, and maintenance costs of both the onsite and offsite leachate treatment scenarios.

| Onsite treatment scenario | | Offsite treatment scenario | | |
|------------------------------|--|---|---|-----------------------|
| Cost sources | Value (in \$2018 USD) | Cost sources | Equation | Value (in \$2018 USD) |
| Capital cost | | Capital cost | | |
| Construction cost | 661,007 ^a | Construction cost of the storage tank | Unit construction cost (\$43.36) ^b × total volume of onsite storage tank (400 m ³) | 17,344 |
| Annual operation cost | | Annual operation and maintenance cost | | |
| Electricity cost | 126,301 ^a | Leachate treatment fee charged by the WWTP (hereafter referred as management fee) | Unit management fee (\$0.18/m ³) ^c × total volume of leachate accepted per year. | 19,412 |
| Chemical cost | 23,841 ^a | | | |
| Maintenance cost | | | | |
| Membrane replacement for MBR | 141,176 ^a (occur in the 5 th , 10 th , and 15 th year) | Leachate transportation cost | \$44/trip ^d × 25 trips/day × 365 days/year | 402,574 |
| Membrane replacement for NF | 145,588 ^a (occur in the 3 th , 6 th , 9 th , 12 th , and 15 th year) | | | |
| Membrane replacement for RO | 139,235 ^a (occur in the 3 rd , 6 th , 9 th , 12 th , and 15 th year) | | | |

^aCost data of the onsite scenario were obtained from report of the Nanjiao landfill

^bUnit construction cost of the offsite scenario was assumed to be the same with the onsite scenario's value which was calculated by dividing onsite scenario's capital cost by total volume of onsite treatment system (15244.7 m³).

^cUnit management fee was obtained from (Hunan Administrative Measures for Collection and Use of Sewage Treatment Fee, 2020).

^dTruck transportation cost per trip was obtained from (Department of Zhuzhou construction cost management, 2018).

2.5. Sensitivity analysis

A sensitivity analysis was conducted to analyze the influence of inputs uncertainty on all environmental and human health impact indicators and economic outcomes for the two leachate treatment scenarios. The tested inputs include construction, electricity and chemical consumptions, membrane replacement, membrane disposal, and direct water emissions. Each of these inputs was varied by ± 20 , ± 50 , and $\pm 100\%$. A sensitivity index (S) was calculated for each input change using Equation 3 (Morris and Fan, 1998).

$$S = \left| \frac{I_i - I_b}{I_b} \right| \times 100\% \quad \text{Equation 3}$$

where, I_i is the altered impact value. I_b is the baseline impact value. Inputs were considered “highly sensitive” if $|S| > 50\%$.

Chapter 3. Results and discussion

The following sections provide the LCA (Section 3.1) and LCCA (Section 3.2) outcomes of the two leachate treatment scenarios under the two FUs, the influence of PFAS concentration on the outcomes (Section 3.3), and the results’ sensitivity to key input parameters (Section 3.4).

3.1. Environmental and human health impact comparison of the two leachate treatment scenarios

Table 4 provides the estimated environmental and human health impacts of the onsite and offsite scenarios under the two FUs. Regardless of the functional unit used, a clear environmental and human health tradeoff exists between the two scenarios. On the basis of 1 m^3 of leachate treated,

shifting leachate treatment from offsite to onsite will increase environmental impacts by 20-23,280%, as a result of higher direct emissions of COD and TSS and higher indirect upstream emissions in the supply chain. On the other hand, treating leachate onsite reduced carcinogenic (HHc) and noncarcinogenic (HHnc) impacts by 67% and 97%, respectively, mainly because of its high PFAS removal efficiencies. On the basis of 1 g of PFAS removed, shifting leachate treatment from offsite to onsite will increase the environmental impacts (except for ET and FFD) by 48-4,859% and decrease the HHc and HHnc impacts by 93% and 99%, respectively. The onsite scenario outperformed the offsite scenario in ET and FFD categories because of the onsite scenario's high PFAS removal efficiency resulted in a larger denominator value (K_{LC}) when 1 g of PFAS was used as a FU. Overall, the offsite scenario performs better environmentally while the onsite scenario performs better from a human health perspective, but the extent of this tradeoff varies depending on the FU used.

Table 4. Environmental and human health result comparisons of the two leachate treatment scenarios under two functional units: 1 m³ of leachate treated and 1 g of PFAS removed. When comparing offsite with onsite leachate treatment scenario, outperformed impact categories are highlighted in green.

| Impact category | Unit | 1 m ³ of leachate treated | | 1 g of PFAS removed | |
|-----------------------------|-----------------------|--------------------------------------|-----------------------|-----------------------|-----------------------|
| | | Onsite scenario | Offsite scenario | Onsite scenario | Offsite scenario |
| Environmental impacts | | | | | |
| Ozone Depletion (OD) | kg CFC11 eq | 1.17×10 ⁻⁴ | 5.00×10 ⁻⁷ | 7.85×10 ⁻⁴ | 1.58×10 ⁻⁵ |
| Global warming (GW) | kg CO ₂ eq | 18.14 | 2.60 | 121.84 | 82.28 |
| Smog (SM) | kg O ₃ eq | 0.86 | 0.10 | 5.78 | 3.19 |
| Acidification (AC) | kg SO ₂ eq | 0.076 | 0.0081 | 0.51 | 0.26 |
| Eutrophication (EU) | kg N eq | 0.020 | 0.0029 | 0.14 | 0.091 |
| Ecotoxicity (ET) | CTUe | 43.14 | 15.75 | 289.72 | 498.63 |
| Fossil fuel depletion (FFD) | MJ surplus | 5.40 | 4.50 | 36.28 | 142.48 |
| Human health impacts | | | | | |
| Carcinogens (HHc) | CTUh | 3.03×10 ⁻⁷ | 9.11×10 ⁻⁷ | 2.03×10 ⁻⁶ | 2.88×10 ⁻⁵ |
| Noncarcinogens (HHnc) | CTUh | 3.04×10 ⁻⁶ | 1.07×10 ⁻⁴ | 2.04×10 ⁻⁵ | 0.0034 |
| Respiratory effects (RE) | kg PM2.5 eq | 0.018 | 0.0017 | 0.12 | 0.055 |

Figure 2 presents the relative contributions of different life cycle phases to each impact indicator for the onsite and offsite scenarios. The high OD impact of the onsite scenario was resulted from the large consumption of MBR membranes (around 99.7% of its total OD impact, Figure 2 (A)). This is because the manufacturing of the PTFE material used for the MBR membranes in the onsite scenario releases a large amount of trifluoroacetic acid into the atmosphere, which can further break to form hydrochlorofluorocarbons and hydrofluorocarbons (Graham, 2001). When the PTFE material is replaced by the PDVF material as used in the offsite scenario, the OD impact of the onsite scenario can be reduced to 4.7×10^{-7} kg CFC11 eq., indicating the importance of using more environmental friendly materials and more effective membrane fouling control (Chen et al., 2018; Ioannou-Ttofa et al., 2016). The onsite scenario's higher GW, SM, AC, EU, and ET impacts were a result of its higher electricity, chemical, and membrane material consumptions during the operation and maintenance phases. They combined contribute to at least 96% of the onsite scenario's GW, SM, AC, EU, and ET impacts (Figure 2 (A)). In comparison, leachate transportation and electricity for wastewater treatment are the major contributors to the environmental impacts of the offsite scenario (Figure 2 (B)). Direct water emissions are the dominant contributor to the HHc (90.6%) and HHnc (99.5%) impacts of the offsite scenario (Figure 2(B)).

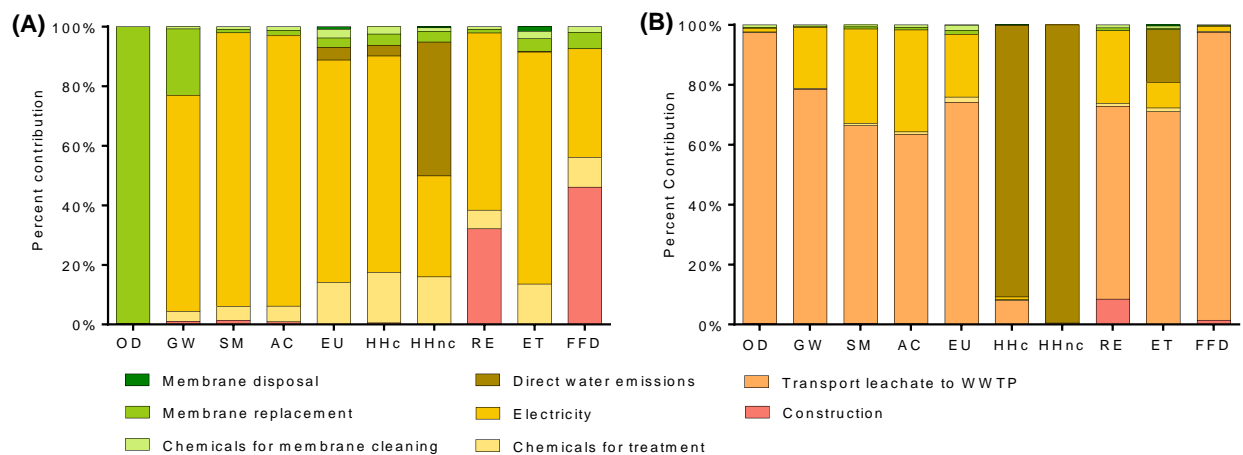


Figure 2. Percent contributions of different life cycle stages to the environmental and human health impacts of the (A) onsite and (B) offsite leachate treatment scenarios. Impact categories abbreviations: ozone depletion (OD), global warming (GW), smog (SM), acidification (AC), eutrophication (EU), carcinogenic (HHc), non-carcinogenic (HHnc), respiratory effects (RE), ecotoxicity (ET), and fossil fuel depletion (FFD).

3.2. Life cycle cost comparison of the two leachate treatment scenarios

Figure 3 compares the total life cycle cost of the two scenarios using both FUs. The life cycle cost to treat 1 m³ of raw leachate is \$1.96 for the onsite scenario and \$2.50 for the offsite scenario. Landfills operators are expected to experience a 21% decrease in life cycle cost when shifting from the offsite to the onsite leachate treatment, despite the onsite scenario's higher initial cost. When using 1 g of PFAS treated as the FU, the onsite scenario's life cycle cost will be 83% lower than the offsite scenario's. Operation of the onsite treatment plant has the highest contribution (45%) to the onsite scenario's life cycle cost, mainly due to the high electricity and chemical usages. In comparison, 95% of the offsite scenario's life cycle cost stems from leachate transportation due the number of trips needed every day. This finding aligns with two previous studies, both of which found that transportation has the highest contribution to the life cycle cost when leachate was treated in WWTPs (Robertson et al., 1995; Ye et al., 2014).

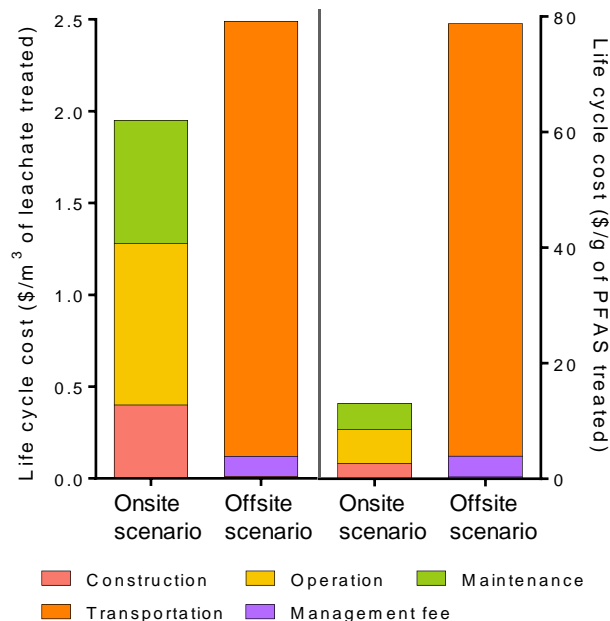


Figure 3. Life cycle costs of the onsite and offsite leachate treatment scenarios under the two studied functional units. Life cycle cost is expressed in \$2018 USD.

3.3. Influence of PFAS concentration on the environmental, human health, and economic tradeoffs

Results obtained from the previous sections show the onsite scenario offers HHc, HHnc, and economic benefits as compared to the offsite scenario, but may result in higher environmental impacts under a raw leachate PFAS concentration of 150,407 ng/L. Figure 4 presents the influence of PFAS concentration on the results using 1 m³ of leachate treated as a FU. Under this FU, PFAS concentration will only influence the ET, HHc, and HHnc outcomes as they are linearly related to the amount of PFAS released to the environment. The offsite scenario outperforms the onsite scenario in terms of ET, HHc, and HHnc when the PFAS concentration is lower than 1.65 mg/L, 38,279 ng/L, and 1,666 ng/L, respectively. Therefore, when PFAS concentration in the raw leachate is less than 1,666 ng/L, the onsite scenario does not offer either environmental or human health benefits, making the shift from offsite to onsite scenario unfavorable.

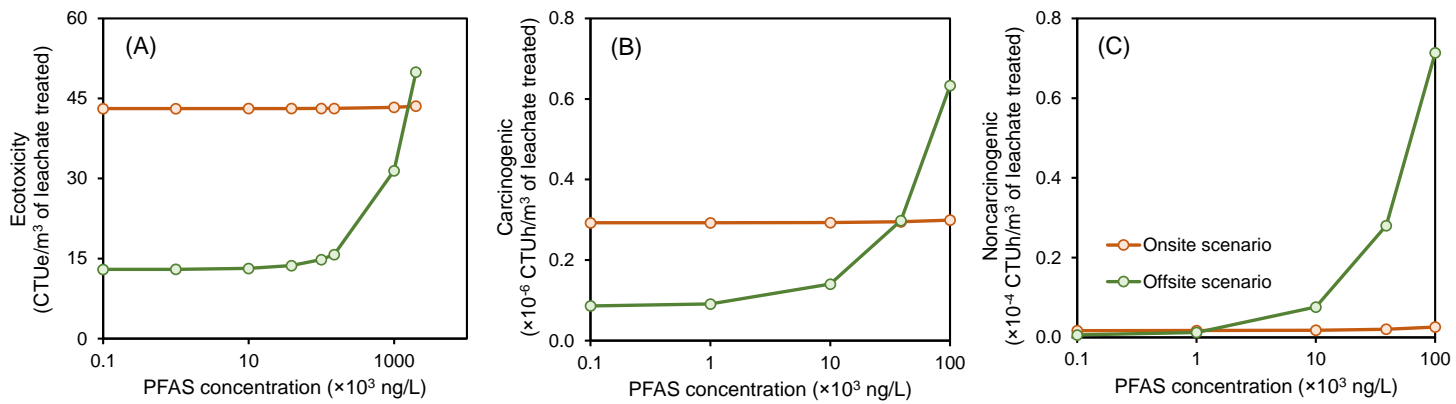


Figure 4. Influence of PFAS concentration on ecotoxicity impacts (A), human health carcinogenic (B) and noncarcinogenic (C) impacts in treating 1 m³ of leachate.

On the other hand, PFAS concentration in the raw leachate will influence all impact outcomes when 1 g of PFAS removed is used as a FU. Figure 5 illustrates such influences using OD, ET, HHc, and the life cycle costs as examples. All studied impacts exhibit an exponential approach pattern with the increase of PFAS concentration. Unit impacts decrease rapidly when PFAS concentration in the raw leachate is relatively low, and gradually approach zero when PFAS concentration keeps increasing. The two scenarios' PFAS mass-based performances converge at around 10,000 ng/L for all impact categories.

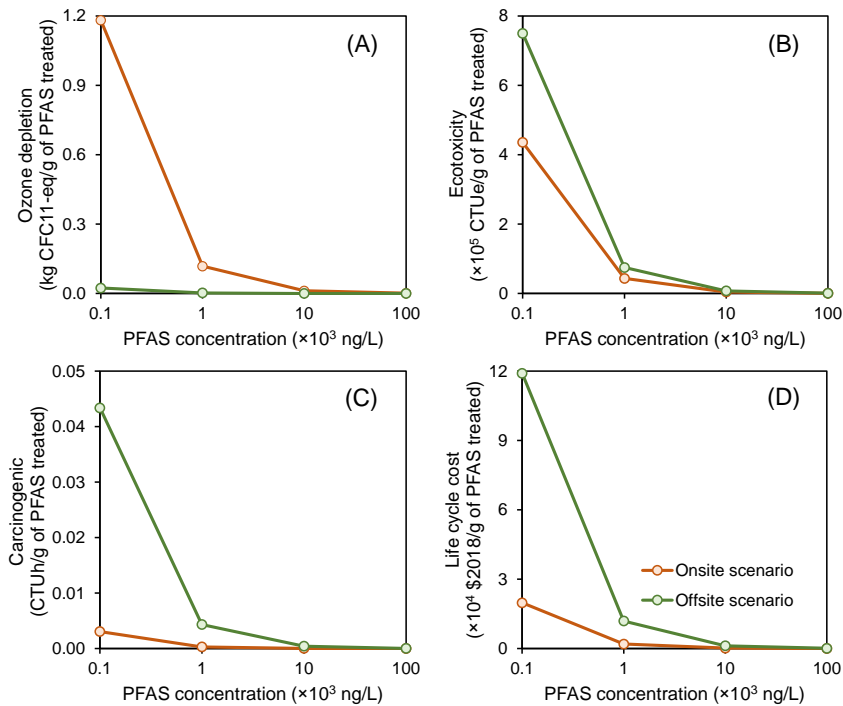


Figure 5. Influence of PFAS concentration on ozone depletion (A), ecotoxicity (B), carcinogenic human health impact (C), and life cycle cost (D) in removing 1 g of PFAS from leachate.

3.4. Sensitivity analysis

The changes of life cycle environmental, human health, and economic implications for the onsite and offsite treatment scenarios in response to changes in key inventory inputs are available in Table S3 and Table S4 in the Appendix, respectively. For the onsite

treatment scenario, its OD impact is highly sensitive to >50% increase or <50% decrease in the number of membranes. All environmental and human health impact categories, except for OD and HHnc, are solely sensitive to significant increases or decreases in electricity consumption. The onsite scenario's life cycle cost is sensitive to large changes in the amount of chemicals used for operation and maintenance. The high sensitivity of onsite scenario's environmental, human health, and economic performances to membrane replacement, electricity and chemical usages indicates the importance of using more effective membrane fouling control, improving energy and chemical usage efficiencies, or applying renewable energy sources. In terms of the offsite scenario, all environmental, human health, and economic impact categories, except for HHc and HHnc, are merely sensitive to $\pm 100\%$ changes in the transportation of leachate from onsite to offsite. The HHc and HHnc impacts are exclusively sensitive to $\pm 100\%$ changes in the mass of direct water emission. Thus, reducing impacts from leachate transportation and direct water emission plays a key role in improving the offsite scenario's performances.

Chapter 4. Conclusion

In light of the increasingly stringent regulations and concerns related to PFAS discharge into the environment, many landfills are under the pressure to shift from offsite to onsite for leachate treatment. Our study of a typical offsite and a typical onsite leachate treatment scenario found that the onsite scenario offers benefits from human health and economic perspectives, while the offsite scenario performs general better environmental outcomes. The extent of this tradeoff varies when different functional units were adopted. If the volume (1 m^3 of leachate treated) was used as the functional unit, shifting offsite

leachate treatment to onsite increases environmental impacts by 20-23,280%, but decreases human health impacts by 67-97% and reduces life cycle cost by 21%. If the PFAS reduction (1 g of PFAS removed) is used as the functional unit, this shifting of leachate treatment results in increasement of environmental impacts (48-4,859%, except for ecotoxicity and fossil fuel depletion) and reduction of ecotoxicity and fossil fuel depletion (42-75%), human health (93-99%), and economic (83%) impacts. Variations of PFAS concentrations in raw leachate may further influence environmental and human health performances of the onsite and offsite scenarios. Our results suggest that the onsite scenario does not offer either environmental or human health benefits when PFAS concentration in the raw leachate is less than 1,666 ng/L, making the shift from offsite to onsite scenario unfavorable. To further improve sustainability of the onsite treatment, it is critical to reduce consumptions of membrane, electricity, and chemical usages of the system.

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Appendix

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Table S1. The construction cost in 2018 and 2002.

| | Construction cost (\$2018) | Consumer price index | Construction cost (\$2002) |
|------------------|----------------------------|----------------------|----------------------------|
| Onsite scenario | 681,492 | 0.72 | 488,001 |
| Offsite scenario | 17,879 | 0.72 | 12,796 |

Table S2. The list of PCBs that used to calculate human health impacts.

| Name of PCBs |
|--|
| Polychlorinated biphenyl, PCB-1016 |
| Polychlorinated biphenyl, PCB-1254 |
| Polychlorinated biphenyl, PCB-1260 |
| Chloro-1,1-biphenyl, PCB-1254 |
| 2,5,2'-Trichlorobiphenyl, PCB-18 |
| 2,3,3',4,4',6-Hexachlorobiphenyl, PCB 158 |
| 2,3,3',4,4'-Pentachlorobiphenyl, PCB 105 |
| 2,3,3',4',6-Pentachlorobiphenyl, PCB 110 |
| 2,3,3',4',5,6-Hexachlorobiphenyl, PCB 160 |
| 2,3',4',5-Tetrachlorobiphenyl, PCB 70 |
| 2,3',4,4',5'-Pentachlorobiphenyl, PCB 123 |
| 2,3',4,4',5-Pentachlorobiphenyl, PCB 118 |
| 2,2',3,3',4,4',5,5'-Octachlorobiphenyl, PCB 194 |
| 2,2',3,3',4,5,5',6'-Octachlorobiphenyl, PCB 199 |
| 2,2',3,4',5',6-Hexachlorobiphenyl, PCB 149 |
| 2,2',3,4,4',5'-Hexachlorobiphenyl, PCB 138 |
| 2,2',3,4,4',5,5'-Heptachlorobiphenyl, PCB 180 |
| 2,3-dichlorobiphenyl, PCB 5 |
| 2,2',5,5'-Tetrachlorobiphenyl, PCB 52 |
| 1,1'-Biphenyl, 4,4'-dichloro-, PCB-15 |
| 1,1'-Biphenyl, 2-chloro-, PCB-1 |
| 1,1'-Biphenyl, 2,2',4,4'-tetrachloro-, PCB-47 |
| 1,1'-Biphenyl, 2,2',4,4',6,6'-hexachloro-, PCB-155 |
| 1,1'-Biphenyl, 2,2',5-trichloro-, PCB-18 |
| 1,1'-Biphenyl, 2,4-dichloro-, PCB-7 |

1,1'-Biphenyl, 2,4,5-trichloro-, PCB-29

1,1'-Biphenyl, 3-chloro-, PCB-2

1,1'-Biphenyl, 3,3',4,4'-tetrachloro-, PCB-77

1,1'-Biphenyl, 4-chloro-, PCB-3

Table S3. Results of sensitivity analysis of the major inventories in the onsite treatment scenario.

| Input | Change (%) | Output | | | | | | | | | | |
|--------------|------------|--------|---------------|---------------|---------------|---------------|---------------|--------|---------------|---------------|---------------|-----------------|
| | | OD | GW | SM | AC | EU | HHc | HHnc | RE | ET | FFD | Life cycle cost |
| Construction | -100 | 0.03% | 0.23% | 0.04% | 0.05% | 0.04% | 0.39% | 0.81% | 0.01% | 0.57% | 2.69% | 8.93% |
| | -50 | 0.01% | 0.11% | 0.02% | 0.03% | 0.02% | 0.20% | 0.40% | 0.01% | 0.29% | 1.35% | 4.46% |
| | -20 | 0.01% | 0.05% | 0.01% | 0.01% | 0.01% | 0.08% | 0.16% | 0% | 0.11% | 0.54% | 1.79% |
| | 20 | 0.01% | 0.05% | 0.01% | 0.01% | 0.01% | 0.08% | 0.16% | 0% | 0.11% | 0.54% | 1.79% |
| | 50 | 0.01% | 0.11% | 0.02% | 0.03% | 0.02% | 0.20% | 0.40% | 0.01% | 0.29% | 1.35% | 4.46% |
| | 100 | 0.03% | 0.23% | 0.04% | 0.05% | 0.04% | 0.39% | 0.81% | 0.01% | 0.57% | 2.69% | 8.93% |
| Electricity | -100 | 0.13% | 73.05% | 93.22% | 91.64% | 74.70% | 72.75% | 33.64% | 87.68% | 77.49% | 66.14% | 16.56% |
| | -50 | 0.07% | 36.52% | 46.61% | 45.82% | 37.35% | 36.37% | 16.82% | 43.84% | 38.74% | 33.07% | 8.28% |
| | -20 | 0.03% | 14.61% | 18.64% | 18.33% | 14.94% | 14.55% | 6.73% | 17.54% | 15.50% | 13.23% | 3.31% |
| | 20 | 0.03% | 14.61% | 18.64% | 18.33% | 14.94% | 14.55% | 6.73% | 17.54% | 15.50% | 13.23% | 3.31% |
| | 50 | 0.07% | 36.52% | 46.61% | 45.82% | 37.35% | 36.37% | 16.82% | 43.84% | 38.74% | 33.07% | 8.28% |
| | 100 | 0.13% | 73.05% | 93.22% | 91.64% | 74.70% | 72.75% | 33.64% | 87.68% | 77.49% | 66.14% | 16.56% |
| Chemicals | -100 | 0.17% | 4.18% | 5.64% | 6.46% | 16.86% | 19.50% | 17.09% | 10.59% | 15.95% | 21.40% | 59.49% |
| | -50 | 0.08% | 2.09% | 2.82% | 3.23% | 8.43% | 9.75% | 8.54% | 5.30% | 7.98% | 10.70% | 29.75% |
| | -20 | 0.03% | 0.84% | 1.13% | 1.29% | 3.37% | 3.90% | 3.42% | 2.12% | 3.19% | 4.28% | 11.90% |
| | 20 | 0.03% | 0.84% | 1.13% | 1.29% | 3.37% | 3.90% | 3.42% | 2.12% | 3.19% | 4.28% | 11.90% |
| | 50 | 0.08% | 2.09% | 2.82% | 3.23% | 8.43% | 9.75% | 8.54% | 5.30% | 7.98% | 10.70% | 29.75% |

| | | | | | | | | | | | | |
|-----------------------|------|---------------|--------|-------|-------|--------|--------|--------|--------|--------|--------|---------------|
| | 100 | 0.17% | 4.18% | 5.64% | 6.46% | 16.86% | 19.50% | 17.09% | 10.59% | 15.95% | 21.40% | 59.49% |
| Direct water emission | -100 | 0% | 0% | 0% | 0% | 4.28% | 3.54% | 44.62% | 0% | 0.08% | 0% | X |
| | -50 | 0% | 0% | 0% | 0% | 2.14% | 1.77% | 22.34% | 0% | 0.04% | 0% | X |
| | -20 | 0% | 0% | 0% | 0% | 0.86% | 0.71% | 8.98% | 0% | 0.02% | 0% | X |
| | 20 | 0% | 0% | 0% | 0% | 0.85% | 0.71% | 8.85% | 0% | 0.02% | 0% | X |
| | 50 | 0% | 0% | 0% | 0% | 2.14% | 1.77% | 22.21% | 0% | 0.04% | 0% | X |
| | 100 | 0% | 0% | 0% | 0% | 4.27% | 3.54% | 44.49% | 0% | 0.08% | 0% | X |
| Membrane replacement | -100 | 99.67% | 22.54% | 1.11% | 1.85% | 3.29% | 3.94% | 6.30% | 1.71% | 4.36% | 9.75% | 15.01% |
| | -50 | 49.83% | 11.27% | 0.55% | 0.93% | 1.64% | 1.97% | 3.15% | 0.85% | 2.18% | 4.87% | 7.51% |
| | -20 | 19.93% | 4.51% | 0.22% | 0.37% | 0.66% | 0.79% | 1.26% | 0.34% | 0.87% | 1.95% | 3.00% |
| | 20 | 19.93% | 4.51% | 0.22% | 0.37% | 0.66% | 0.79% | 1.26% | 0.34% | 0.87% | 1.95% | 3.00% |
| | 50 | 49.83% | 11.27% | 0.55% | 0.93% | 1.64% | 1.97% | 3.15% | 0.85% | 2.18% | 4.87% | 7.51% |
| | 100 | 99.67% | 22.54% | 1.11% | 1.85% | 3.29% | 3.94% | 6.30% | 1.71% | 4.36% | 9.75% | 15.01% |
| Membrane disposal | -100 | 0% | 0.01% | 0% | 0% | 1.01% | 0.02% | 0.64% | 0% | 1.55% | 0% | X |
| | -50 | 0% | 0% | 0% | 0% | 0.51% | 0.01% | 0.32% | 0% | 0.77% | 0% | X |
| | -20 | 0% | 0% | 0% | 0% | 0.20% | 0.00% | 0.13% | 0% | 0.31% | 0% | X |
| | 20 | 0% | 0% | % | 0% | 0.20% | 0.00% | 0.13% | 0% | 0.31% | 0% | X |
| | 50 | 0% | 0% | 0% | 0% | 0.51% | 0.01% | 0.32% | 0% | 0.77% | 0% | X |
| | 100 | 0% | 0.01% | 0% | 0% | 1.01% | 0.02% | 0.64% | 0% | 1.55% | 0% | X |

Table S4. Results of sensitivity analysis of the major inventories in the offsite treatment scenario.

| Input | Change (%) | Output | | | | | | | | | | |
|--------------|------------|--------|-------|-------|-------|-------|------|------|-------|------|------|-----------------|
| | | OD | GW | SM | AC | EU | HHc | HHnc | RE | ET | FFD | Life cycle cost |
| Construction | -100 | 0.3% | 0.2% | 0.3% | 0.2% | 0.0% | 0.0% | 0.0% | 8.4% | 0.0% | 1.4% | 0.4% |
| | -50 | 0.2% | 0.1% | 0.2% | 0.1% | 0.0% | 0.0% | 0.0% | 4.2% | 0.0% | 0.7% | 0.2% |
| | -20 | 0.1% | 0.0% | 0.1% | 0.0% | 0.0% | 0.0% | 0.0% | 1.7% | 0.0% | 0.3% | 0.1% |
| | 20 | 0.1% | 0.0% | 0.1% | 0.0% | 0.0% | 0.0% | 0.0% | 1.7% | 0.0% | 0.3% | 0.1% |
| | 50 | 0.2% | 0.1% | 0.2% | 0.1% | 0.0% | 0.0% | 0.0% | 4.2% | 0.0% | 0.7% | 0.2% |
| | 100 | 0.3% | 0.2% | 0.3% | 0.2% | 0.0% | 0.0% | 0.0% | 8.4% | 0.0% | 1.4% | 0.4% |
| Electricity | -100 | 1.3% | 20.4% | 31.5% | 34.1% | 20.9% | 1.0% | 0.0% | 24.3% | 8.5% | 1.8% | X |
| | -50 | 0.6% | 10.2% | 15.8% | 17.0% | 10.5% | 0.5% | 0.0% | 12.1% | 4.2% | 0.9% | X |
| | -20 | 0.3% | 4.1% | 6.3% | 6.8% | 4.2% | 0.2% | 0.0% | 4.9% | 1.7% | 0.4% | X |
| | 20 | 0.3% | 4.1% | 6.3% | 6.8% | 4.2% | 0.2% | 0.0% | 4.9% | 1.7% | 0.4% | X |
| | 50 | 0.6% | 10.2% | 15.8% | 17.0% | 10.5% | 0.5% | 0.0% | 12.1% | 4.2% | 0.9% | X |
| | 100 | 1.3% | 20.4% | 31.5% | 34.1% | 20.9% | 1.0% | 0.0% | 24.3% | 8.5% | 1.8% | X |
| Chemicals | -100 | 1.1% | 0.8% | 1.4% | 1.8% | 3.5% | 0.4% | 0.0% | 2.0% | 2.0% | 0.5% | X |
| | -50 | 0.6% | 0.4% | 0.7% | 0.9% | 1.8% | 0.2% | 0.0% | 1.0% | 1.0% | 0.2% | X |
| | -20 | 0.3% | 0.2% | 0.4% | 0.5% | 0.9% | 0.1% | 0.0% | 0.5% | 0.6% | 0.1% | X |
| | 20 | 0.3% | 0.2% | 0.4% | 0.5% | 0.9% | 0.1% | 0.0% | 0.5% | 0.6% | 0.1% | X |
| | 50 | 0.6% | 0.4% | 0.7% | 0.9% | 1.8% | 0.2% | 0.0% | 1.0% | 1.0% | 0.2% | X |

| | | | | | | | | | | | | | |
|-----------------------|----------------------|------|------|------|------|------|--------------|--------------|------|-------|------|------|---|
| Direct water emission | 100 | 1.1% | 0.8% | 1.4% | 1.8% | 3.5% | 0.4% | 0.0% | 2.0% | 2.0% | 0.5% | X | |
| | -100 | 0.2% | 0.4% | 0.7% | 0.8% | 1.4% | 90.7% | 99.5% | 0.9% | 18.2% | 0.3% | X | |
| | -50 | 0.2% | 0.4% | 0.7% | 0.8% | 1.3% | 45.4% | 49.8% | 0.9% | 9.3% | 0.3% | X | |
| | -20 | 0.2% | 0.4% | 0.7% | 0.8% | 1.3% | 18.2% | 19.9% | 0.9% | 4.0% | 0.3% | X | |
| | 20 | 0.2% | 0.4% | 0.7% | 0.8% | 1.3% | 18.2% | 19.9% | 0.9% | 4.0% | 0.3% | X | |
| | 50 | 0.2% | 0.4% | 0.7% | 0.8% | 1.3% | 45.4% | 49.8% | 0.9% | 9.3% | 0.3% | X | |
| | 100 | 0.2% | 0.4% | 0.7% | 0.8% | 1.4% | 90.7% | 99.5% | 0.9% | 18.2% | 0.3% | X | |
| | Membrane replacement | -100 | 0.2% | 0.4% | 0.7% | 0.8% | 1.3% | 0.1% | 0.1% | 0.9% | 0.5% | 0.3% | X |
| | | -50 | 0.1% | 0.2% | 0.3% | 0.4% | 0.7% | 0.1% | 0.1% | 0.5% | 0.2% | 0.1% | X |
| | | -20 | 0.0% | 0.1% | 0.1% | 0.2% | 0.3% | 0.1% | 0.1% | 0.2% | 0.1% | 0.1% | X |
| | | 20 | 0.0% | 0.1% | 0.1% | 0.2% | 0.3% | 0.1% | 0.1% | 0.2% | 0.1% | 0.1% | X |
| | | 50 | 0.1% | 0.2% | 0.3% | 0.4% | 0.7% | 0.1% | 0.1% | 0.5% | 0.2% | 0.1% | X |
| | Membrane disposal | 100 | 0.2% | 0.4% | 0.7% | 0.8% | 1.3% | 0.1% | 0.1% | 0.9% | 0.5% | 0.3% | X |
| | | -100 | 0.0% | 0.0% | 0.0% | 0.0% | 0.1% | 0.1% | 0.1% | 0.0% | 0.4% | 0.0% | X |
| -50 | | 0.0% | 0.0% | 0.0% | 0.0% | 0.1% | 0.1% | 0.1% | 0.0% | 0.2% | 0.0% | X | |
| -20 | | 0.0% | 0.0% | 0.0% | 0.0% | 0.0% | 0.1% | 0.1% | 0.0% | 0.1% | 0.0% | X | |
| 20 | | 0.0% | 0.0% | 0.0% | 0.0% | 0.0% | 0.1% | 0.1% | 0.0% | 0.1% | 0.0% | X | |
| 50 | | 0.0% | 0.0% | 0.0% | 0.0% | 0.1% | 0.1% | 0.1% | 0.0% | 0.2% | 0.0% | X | |
| 100 | 0.0% | 0.0% | 0.0% | 0.0% | 0.1% | 0.1% | 0.1% | 0.0% | 0.4% | 0.0% | X | | |

Table S5. The comparison results of TRACI and ReCiPe methods.

| Electricity | | | | | | MBR | | | | | |
|-----------------------|---------------|----------|------------------------------|---------------|----------|-----------------------|---------------|----------|------------------------------|---------------|----------|
| TRACI | | | ReCiPe | | | TRACI | | | ReCiPe | | |
| Categories | Unit | Value | Categories | Unit | Value | Categories | Unit | Value | Categories | Unit | Value |
| Ozone depletion | kg CFC -11 eq | 1.31E-07 | Ozone depletion | kg CFC -11 eq | 5.81E-08 | Ozone depletion | kg CFC -11 eq | 1.17E-04 | Ozone depletion | kg CFC -11 eq | 1.17E-04 |
| Global warming | kg CO2 eq | 1.09E+01 | Climate change | kg CO2 eq | 1.09E+01 | Global warming | kg CO2 eq | 4.01E+00 | Climate change | kg CO2 eq | 4.01E+00 |
| Acidification | kg SO2 eq | 5.68E-02 | Terrestrial acidification | kg SO2 eq | 5.06E-02 | Acidification | kg SO2 eq | 1.32E-03 | Terrestrial acidification | kg SO2 eq | 1.04E-03 |
| Eutrophication | kg N eq | 1.24E-02 | Marine eutrophication | kg N eq | 1.38E-03 | Eutrophication | kg N eq | 6.09E-04 | Marine eutrophication | kg N eq | 3.25E-05 |
| Carcinogenics | CTUh | 1.80E-07 | Freshwater eutrophication | kg P eq | 1.50E-03 | Carcinogenics | CTUh | 1.08E-08 | Freshwater eutrophication | kg P eq | 7.76E-05 |
| Non carcinogenics | CTUh | 8.54E-07 | Human toxicity | kg 1,4-DB eq | 1.24E+00 | Non carcinogenics | CTUh | 8.61E-08 | Human toxicity | kg 1,4-DB eq | 3.55E-01 |
| Respiratory effects | kg PM2.5 eq | 8.72E-03 | Particulate matter formation | kg PM10 eq | 2.28E-02 | Respiratory effects | kg PM2.5 eq | 1.99E-04 | Particulate matter formation | kg PM10 eq | 4.21E-04 |
| Ecotoxicity | CTUe | 2.75E+01 | Terrestrial ecotoxicity | kg 1,4-DB eq | 1.35E-04 | Ecotoxicity | CTUe | 1.81E+00 | Terrestrial ecotoxicity | kg 1,4-DB eq | 1.45E-05 |
| Fossil fuel depletion | MJ surpluses | 1.63E+00 | Freshwater ecotoxicity | kg 1,4-DB eq | 5.31E-02 | Fossil fuel depletion | MJ surpluses | 2.31E-01 | Freshwater ecotoxicity | kg 1,4-DB eq | 2.24E-03 |

| | | | | | | | | | | | |
|------|----------|----------|---------------------------------|--------------|----------|------|----------|----------|---------------------------------|--------------|----------|
| Smog | kg O3 eq | 6.56E-01 | Marine ecotoxicity | kg 1,4-DB eq | 4.92E-02 | Smog | kg O3 eq | 8.39E-03 | Marine ecotoxicity | kg 1,4-DB eq | 2.18E-03 |
| | | | Fossil depletion | kg oil eq | 2.18E+00 | | | | Fossil depletion | kg oil eq | 4.80E-02 |
| | | | Photochemical oxidant formation | kg NMVO C | 3.05E-02 | | | | Photochemical oxidant formation | kg NMVO C | 5.04E-04 |
| | | | Ionising radiation | kBq U235 eq | 1.88E-01 | | | | Ionising radiation | kBq U235 eq | 2.05E-02 |
| | | | Agriculture land occupation | m2a | 1.55E-01 | | | | Agriculture land occupation | m2a | 9.53E-03 |
| | | | Urban land occupation | m2a | 9.18E-02 | | | | Urban land occupation | m2a | 1.58E-03 |
| | | | Natural land transformation | m2 | 4.63E-04 | | | | Natural land transformation | m2 | 3.43E-05 |
| | | | Water depletion | m3 | 1.70E-02 | | | | Water depletion | m3 | 3.24E-03 |
| | | | Metal depletion | kg Fe eq | 6.62E-02 | | | | Metal depletion | kg Fe eq | 1.40E-02 |
