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# Environmental and economic performances of incorporating Fenton-based processes into traditional sludge management systems

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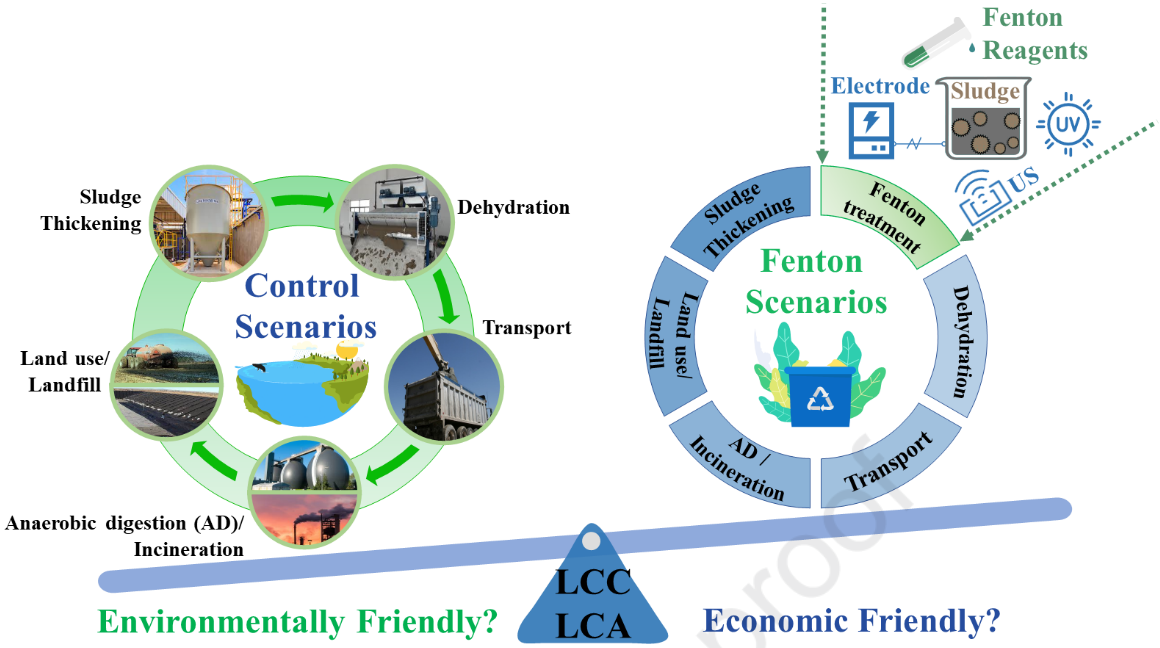
## *Abstract*

Municipal and industrial wastewater treatment plants produce a tremendous amount of sludge containing organic and toxic components. One of the advanced oxidation processes (AOP) - Fenton process has demonstrated great prospect in reduction of sludge organics and toxicity. Fenton pretreatment could ameliorate the sludge dewaterability and biodegradability for anaerobic digestion (AD) process, and enhance the sludge lower heating value for incineration process, thus stimulating sludge dewatering, reduction and energy recovery. However, doubts remain about whether the incorporation of the Fenton process into the traditional sludge management systems brings environmental benefits. Hence, a life cycle environmental impact calculation model was established for sludge with various organic contents (60%, 70%, 80%) under the effect of Fenton and US/UV/Electro-Fenton processes. Noteworthy mitigation of environmental load was observed for the Fenton process coupled with incineration system, which involves high dewatering demand. Conversely, as for the AD system with high biomass transformation rate, Fenton process failed to attain the assumed promotion of environmental benefit. Hydrogen peroxide（$H_2O_2$) prominently attributed to the weakness of Fenton process combined with AD (F-AD) scenario, compared with the AD scenario in terms of environmental impact. Summarily, the F-AD scenario acts as the preponderant system when weighing up the pros and cons of environmental impact, energy balance and life cycle cost. Contrary to the mainstream view, the proven technical advantages of Fenton process cannot compensate for the additional environmental loads in the life cycle of sludge. It provides valuable reflection for environmental managers and scholars that we should be more cautious in the application of cutting-edge technologies.

**Keywords:** Sewage sludge disposal, Fenton process, US/UV/Electro-Fenton processes, Life cycle assessment, Life cycle cost


Sludge
Thickening
Dehydration
Control
Scenarios
Transport
Land use/
Landfill
Anaerobic digestion (AD)/
Incineration
Fenton
Reagents
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Sludge
UV
US
Sludge
Thickening
Fenton
treatment
Fenton
Scenarios
Land use/
Landfill
Dehydration
AD /
Incineration
Transport
LCC
LCA
Environmentally Friendly?
Economic Friendly?

## *Abbreviations*

| | |
|---|---|
| AD | Anaerobic digestion |
| CC | Climate change |
| CHP | Combined heat and power |
| EBR | Energy balance ratio |
| FDP | Fossil depletion potential |
| FEP | Freshwater eutrophication potential |
| FET | Freshwater eco-toxicity |
| GHG | Greenhouse gas |
| HT | Human toxicity |
| IPCC | Intergovernmental panel on climate change |
| IR | Ionizing radiation |
| LCA | Life cycle assessment |
| LCC | Life cycle cost |
| LCI | Life cycle inventory |
| MD | Metal depletion |
| MEP | Marine eutrophication potential |
| MET | Marine eco-toxicity |
| NMVOCs | Non-methane volatile organic compounds |
| ODP | Ozone depletion potential |
| PMF | Particulate matter formation |
| POF | Photochemical oxidation formation |
| TAP | Terrestrial acidification potential |
| TET | Terrestrial eco-toxicity |
| WD | Water depletion |

# Highlights

- Life cycle insights into Fenton processes for sludge were creatively proposed
- Fenton scenarios were favorable in reclamation but not in environmental impacts
- US/UV/electro-Fenton methods have more negative impact due to high energy demand
- Introducing Fenton methods could provoke less life cycle costs in all scenarios
- Alternatives to Fenton reagents could be pressing challenges

## *1. Introduction*

Under the stringent policy requirements, paying equal attention to sludge and water has become the consensus of environmentalists. By the end of 2019, the daily treatment capacity of municipal wastewater treatment plants in China reached 177 million $m^3$/d, with an annual sludge productivity of 39.04 million tons (80% water content) (Wei et al., 2020). Sludge is the solid waste produced during the sewage treatment, and contains plenty of pathogens, heavy metals, organic pollutants, etc., posing potential threats to human health and natural environment, such as odor, disease spread and global warming (Singh et al., 2011). Consequently, it is imminent to implement a series of treatment and disposal procedures, such as thickening, AD, dewatering, thermal drying, incineration, landfill, etc. Sludge disintegration and dewatering are essential steps to maximize the reduction of sludge water content and volume (Wu et al., 2020).

The abundant extracellular polymers (EPS) in sludge absorbs massive water, which conduces to the stable floc structure, and limits the dewatering performance (Mowla et al., 2013). To conquer the above challenges, advanced oxidation process (AOP) emerges. Among them, the degradation of EPS and toxic pollutants into harmless products is easily accomplished by Fenton process, for the nonselective attack from strong oxidizing hydroxyl radicals (OH·). Fenton process has also been universally employed in the industrial wastewater treatment, such as textile (Xavier et al., 2015), medicine (Mackulak et al., 2015), and agricultural chemicals (Saini et al., 2017), as well as the sludge treatment field (Lin et al., 2020). Fenton process involves the reaction between $H_2O_2$ and ferrous iron (catalyst) to produce highly active OH· (Pilli et al., 2015), which is effective for the decomposition of EPS and the microorganisms cell lysis. Furthermore, Fenton process stimulates the release of intracellular substances and bound water (Tony et al., 2008). The possibility of eliminating heavy metals (Yuan et al., 2020) and odor (Liu et al., 2016) in sludge has also been confirmed. Additionally, Fenton process coupled with other methods can further reduce the dosage of Fenton reagent and improve the utilization rate of OH· (Ramteke and Gogate, 2015).

AD, incineration and landfilling are the most common waste sludge management alternatives in developing countries. One of the sludge management dilemmas is the prevention and solution of environmental problems. Life Cycle Assessment (LCA) has been highly prized in the evaluation field of sludge management strategies, as it can quantify the potential impacts from a holistic perspective (Ding et al., 2021). None of existing LCA studies for sludge assess the life cycle impact and cost with respect to preliminary AOP together with traditional disposal (Teoh and Li, 2020). With growing knowledge of "peaking carbon dioxide emissions" and "carbon neutrality" (Ma et al., 2021), environmental cost in life cycle cost (LCC) was born, that is, the cost of environmental pollution and ecological damage (Tarpani and Azapagic, 2018). Nevertheless, most of the LCC studies limited the discussion to private costs (e.g., capital cost, operating cost, transportation cost), without in-depth analysis of the external costs (e.g., external environmental cost) (Woon and Lo, 2016).

Multiple researches have attempted to perceive the optimal conditions for Fenton treatment of sludge, i.e., $H_2O_2$/ iron concentration, reaction time, temperature, and initial pH (Pilli et al., 2015; Zhang et al., 2014). But there are some potential problems in Fenton oxidation, such as large dosage, high cost and strong acidity of filtrate. Fenton reagent may not be an environmentally invasive material, it is undeniable that it can induce certain harm to the environment and human health.

A few studies have thoroughly investigated the life cycle impact raised by those proven techniques for sludge disposal, while merely considering individual aspects of environmental impact or certain process of sludge disposal. To the best of our knowledge, scarcity of environmental evaluation for sludge AOP pretreatment is apparent. This study innovatively incorporated Fenton processes into the full life cycle of sludge to evaluate the trade-off between its performance excellence (dewaterability, heavy metal and odor elimination, etc.) and environmental risk or cost (additional material and energy input). To this end, the ambitions of this study were: (i) to supplement LCA and LCC data gaps for Fenton-pretreated sludge of various organic contents; (ii) to execute systematic research on the application of Fenton process for enhancing sludge dewatering and reduction, including the selection of Fenton methods and the matching between dosage and sludge disposal routes, etc.; (iii) to scrutinize the energy efficiencies of Fenton-pretreated sludge with various organic contents. The procedure that combines scenario storylines and quantitative assessment was reasonably used, based on empirical laboratory tests and in-situ monitoring data in complementary roles. Here, the multi-dimensional results deliver important insights into the source, quality and occurrence law of environmental loads, as well as guidance of green development, when sludge characteristics are optimized via Fenton processes.

## *2. Materials and methods*

### *2.1 Framework of LCA*

One ton of dry solid (1t DS) was set as the functional unit (FU) in this study, that is, 100 tons of concentrated sludge containing 1t DS (moisture content 99%). The conversions of material, energy and pollutant input and output in all processes corresponded to the treatment for 1t DS. And all the data diagrams were completed by Origin 2018.

#### *2.1.1 System boundary and scenario descriptions*

In this study, the system boundary was defined as following six systems：

(1) F-L: Sludge thickening, Fenton pretreatment, Dewatering, Landfill

(2) F-AD: Sludge thickening, Fenton pretreatment, Anaerobic digestion (AD), Dewatering, Land use

(3) F-I: Sludge thickening, Fenton pretreatment, Dewatering, Thermal drying, Incineration, Landfill

(4) EF-AD: Sludge thickening, Electro-Fenton pretreatment, AD, Dewatering, Land use

(5) US+F-AD: Sludge thickening, Ultrasound-Fenton pretreatment, AD, Dewatering, Land use

(6) UV+F-AD: Sludge thickening, Ultraviolet-Fenton pretreatment, AD, Dewatering, Land use

**Fig. 1**

To delve into the intermediate unit processes with considerable contribution, and explore the dominant input variables in each unit process, the processes of sludge dewatering, reduction, transportation and final disposal were listed as the system boundary. For each system, a control scenario was set, which referred to the sludge treatment and disposal routes without Fenton pretreatment.

Considering the comparability and universality for different routes, following assumptions and limitations were adopted within the system boundary, based on the acknowledged setting and cut-off criteria in existing LCA results (Gentil et al., 2010):

(1) The sewage treatment line was uniform in all scenarios, and thus not included in the system.

(2) This study targets at sludge management, thus residual sludge was selected as the input material, with an assumption of different organic contents (60, 70, and 80%) in AD and incineration scenarios.

(3) The construction of plants, vehicles, machinery and auxiliary equipment were excluded in the system boundary, with an assumption of transportation distance of 30 km in all cases (Yoshida et al., 2013).

(4) Biogas supplied by anaerobic digestion and waste heat from incineration was applied in combined heat and power (CHP) generation.

(5) Carbon dioxide from the biomass decomposition in landfills as well as burning of biogas in CHP and dried sludge in incinerators, were deemed to be carbon neutral (biological source).

The system boundary is shown in Fig. 1. The energy (electricity, natural gas, coal and diesel) and materials (polymer, $H_2O_2$, $FeSO_4$, etc.), as well as the residual power output and pollutant emissions are shown in Table S1-1 ~ Table S1-4. Renewable power will replace partial power grid, and the substitution effect was calculated with reference to national data. The waste water from dewatering and drying process was conveyed to the sewage treatment plant, taking transportation power and treatment cost into account.

In the landfill route (widely applied in China), the sludge was transported to the landfill sites by truck and dumped into the ditch from the vehicle. High density polyethylene (HDPE) membranes were used for landfill site covering and anti-seepage. Lime was added to the dewatered sludge for stabilization.

In the AD route, the concentrated sludge was heated to 35-40°C before entering the mixed digestion tank, with electricity recovery efficiency of 34% and thermal energy recovery efficiency of 40%. After digestion, the digestate was dehydrated into sludge cake, then transported and scattered on land for reuse. The effect of fertilizer substitution was not considered in this study.

In the incineration route, the sludge was burned in the incinerator after pre-drying processes like dewatering and thermal drying. Existing studies have validated that the incineration slag and fly ash usually end up in the landfill (Nakatsuka et al., 2020; Yoshida et al., 2018). For the landfill with reliable function, it is assumed that the sludge ash would not release heavy metals and other pollutants to the environment.

### *2.1.2 Life cycle inventory analysis*

The life cycle inventory (LCI) data in this study were principally collected from the field and experimental data in various literatures and industrial reports issued by governments. The background data were complemented with the global or European average production data in the Ecoinvent database. See supplementary materials for details.

## 2.2 Analysis and evaluating system

### 2.2.1 Environmental impact assessment

The environmental impact assessment relied on the openLCA 1.10.2 software (https://openlca.org), ReCiPe Midpoint (E) method and educational Ecoinvent 3.7.1 database (https://nexus.openlca.org/database/ecoinvent) embedded in this software. The environmental impact of Fenton-pretreated sludge on 15 categories of environmental damage and the total impact potential were analyzed. The characterization results were normalized to obtain dimensionless numbers, i.e., the ratio of each characterization results to the normalization factor of Recipe (Table S5).

### 2.2.2 Uncertainty analysis and sensitivity analysis

Uncertainty analysis was employed to convincingly support the conclusion. Data quality indexes (Weidema and Wesnæs, 1996) was coupled with the random quantitative method for qualitative and quantitative analyses of data quality. First, contribution analysis was utilized to screen all the results and then ignore the less important impact categories and processes. The second step was to determine the probability distribution function by scrutinizing the uncertainty lists proposed by Clavreul et al. (2012), which were commonly used in LCA of solid waste. Thirdly, the Monte Carlo simulation (Eggleston et al., 2006) recommended in the guidelines of the Intergovernmental Panel on Climate Change (IPCC) was used to quantify the uncertainty of assessment results, originating from the statistical variability of LCI data. Based on the uncertainty of LCI data represented by probability distribution, the Monte Carlo function established in openLCA ran 10,000 iterations at significance level of 0.05 to present the uncertainty propagation results.

### 2.2.3 Energy efficiency analysis

Energy balance ratio (EBR) stands for the ratio of sludge energy recovered in the form of biogas or incineration heat to net energy input (electricity, heat, thermal power of fossil fuel), partly reflecting the energy cost of recovering energy from sludges. EBR=Eout / Ein. Ein means the external energy requisited for the system, and Eout is the output energy of the system. When EBR exceeds 1, the system can achieve energy self-sufficiency.

### *2.2.4 Life cycle cost analysis*

LCC of sludge management strategies refers to the environmental damage caused by greenhouse gas (GHG) emissions (including direct emissions and indirect offsetting emissions) in different scenarios, as well as all economic costs of the whole system (Jamasb and Nepal, 2010). The LCC was comprised of two parts: economic cost (Francini et al., 2019) and environmental cost (Woon and Lo, 2016). The LCC analysis was completed in the following order: select target facilities, investigate basic assumptions (such as discount rate and carbon trading cost), classify costs (such as investment cost and operating cost), investigate relevant market data, and measure the time value of money. See section S4 of supplementary materials for the detailed calculation process.

## *3. Results*

### *3.1 Environmental impacts for Fenton-pretreated sludge*

### *3.1.1 Landfill scenario*

In terms of all the midpoint impact categories, the conventional landfill route combined with Fenton-pretreatment was higher than the control (Fig. 2), indicating that Fenton process failed to bring enough environmental sustainability. For the above two scenarios, they both showed significant impacts in the normalized results for climate change (CC), fossil depletion potential (FDP), freshwater eutrophication potential (FEP) and marine eutrophication potential (MEP). In the control (Landfill) scenario, above significant impacts were respectively attributed to the greenhouse effect (61%), PAM input (41%), power consumption (48%) and landfill gas

(58%). For the F-L scenario, that were mainly ascribed to the Fenton reagent. Contribution from input substances were shown below: CC ($H_2O_2$ accounted for 38%, quicklime accounted for 31%), FRD ($H_2O_2$ 46%, $H_2SO_4$ 14%), FEP ($H_2O_2$ 56%), MEP (landfill gas 35%, $H_2O_2$ 24%). A larger distinction in ET and human toxicity (HT) categories was observed between F-L scenario and the control, for the long-term impact caused by the direct discharge of heavy metals during the production of $H_2O_2$ by anthraquinone process. In the control scenario, the prominent contribution derived from the landfill process. Whereas, the Fenton process astonishingly exceeded the contribution from the landfill process, being the largest contributor for most impact indicators (Fig. S2-1). The key substances in the Fenton treatment process were identified in Fig. S2-2, i.e., $H_2O_2$ > CaO > $H_2SO_4$ > $FeSO_4$. Virtually, the harsh pretreatment conditions of Fenton process restrict its practical engineering application, and introduce the unexpected environmental impact.

**Fig. 2**

### *3.1.2 Anaerobic digestion scenario for high organic sludge*

For the F-AD scenarios with three high levels of organic contents (60%, 70% and 80%), similar results were found in terrestrial acidification potential (TAP), photochemical oxidation formation (POF) and MEP categories (Fig. 3). With the increase of sludge organics, both of the acid gas emissions from AD process and avoided emissions from CHP marginally increased. Therefore, the decline of acidification potential with high organic content was limited, so were POF and MEP. The normalized results imply that the pivotal environmental threats of F-AD scenario derived from TAP, POF and particulate matter formation (PMF). Additionally, the emission of $NO_X$ and $SO_X$ during AD, and the increased consumption of $H_2SO_4$ vastly contributed to them. 21.8%, 23.9% and 67.7% reduction for the above three impacts were observed in sludge with organic content of 80%, compared with that of 60%.

**Fig. 3**

The increased biogas yield (20%~30%) exerted by the Fenton reagent was incapable of offsetting the extra environmental load in any impact category, even under the feedstock with the highest organic content (Fig. S2-3). The differences were mainly observed in TAP, PMF, POF, terrestrial eco-toxicity (TET) and CC, resulting from the emission of $SO_2$, $NO_X$ and heavy metals (Cu, Zn, Ni, etc.) during the production of $H_2SO_4$ and $H_2O_2$.

The AD process could be identified as a key process (Fig. S2-4), with a negative contribution (~94%). On the other hand, the power, PAM and diesel consumption for sludge concentration, dewatering and transportation delivered significant positive contribution. While in F-AD scenario, what stood out was the phenomenal positive contribution from Fenton process (28%~65%). And compared with the control, the negative contribution brought by AD process declined (-5%~58%).

### *3.1.3 Incineration scenario for high organic sludge*

Fenton pre-oxidation poses a positive impact on the mechanical dewatering and drying characteristics (e.g., calorific value) of dewatered sludge (Liu et al., 2014; Menon et al., 2020). The solid content of sludge cake can even reach 40% under the vacuum pressure of 0.5 bar (Dewil et al., 2005).

All impact indicators of sludge incineration scenarios with three levels of organic contents are shown in Fig. 4. Evidently, all indicators for F-I-80% scenario fell to lowest point, with the minimum impact on metal depletion (MD), TET and ozone depletion potential (ODP), and the maximum impact on MEP, FEP and TAP. The incorporation of Fenton process could significantly reduce the impact on CC, ODP, FDP, HT and ionizing radiation (IR), chiefly due to the heat and power recovery. However, more resources (i.e., chemicals for Fenton reaction, acidification and subsequent neutralization) are demanded to achieve such a reduction, which reversely aggravates environmental risks like MD, water depletion (WD) and MEP.

Fenton-pretreated sludge with three levels of organics exhibited similar environmental impact in the incineration scenario (Fig. 4), yielding 9% ~ 32% contrast in CC, freshwater eco-toxicity (FET) and MD categories. The difference in CC stemmed from chemical production and fossil fuel, and the ecotoxicity was induced by the leachate emission into soil and atmosphere. The environmental benefits brought by the increase of sludge organic content and calorific value would be submerged by heavy metal pollution (Li et al., 2017).

Via Fenton-pretreatment, the reduced moisture content of dewatered sludge met the requirement of self-sustaining combustion, omitting unnecessary thermal drying process and motivating all-round reduction of various environmental impact, especially in CC and ODP (Fig. S2-5). Figure S2-6 reveals that in the control scenario, thermal drying process were primarily responsible for the impact, followed by incineration process. In the F-I scenario, Fenton pretreatment process became an important driving factor, averagely accounting for more than 50% of six impact categories.

**Fig. 4**

### *3.1.4 Comparative analysis*

Conclusively, the rank of environmental impact potential of each scenario was as follows: Incineration > F-I > F-L > landfill > F-AD > AD (Table S2-2, S2-4, S2-6). For landfill route, the total impact potential of F-L scenario was 2.3 times that of the control. With respect to AD route for sludge with three levels of organics, F-AD scenario aroused an environmental benefit reduction of 125% ~ 35%, hence indicating there was a weak link between the Fenton pretreatment and expected improvement in environmental benefits. Regarding incineration route, the introduction of Fenton process led to approximately 47%~48% reduction of environmental impact, but unavailable for positive environmental benefit.

## 3.2 Environmental impact for US/UV/Electro-Fenton pretreated sludge with AD scenario

Sludge reduction and stabilization with high organic generally require more chemicals and energy to fulfill the requirements of disposal. High organic content also portends high biomass and energy recovery potential (Chen et al., 2022). Fenton process combined with other methods succeed in further strengthening sludge dewatering, reducing the environmental hazards, and enhancing subsequent AD efficiency for high organic sludge (Feki et al., 2020; Heng et al., 2017; Ning et al., 2014). Therefore, AD performance for US/UV/Electro-Fenton pretreated sludge with high organics was investigated in particular.

### 3.2.1 EF-AD scenario

The total impact dropped with the increase of organic contents (55% and 29%, respectively) (Table S2-8). While all the positive environmental impact demonstrated that the increased biogas production from high organic sludge could not offset the energy input for the whole system. It is worth noting that under 70% and 80% organic contents, there were negative contributions in terms of CC, FRD and ODP, implying strong correlation between organic content and above impact categories. FEP (21% ~ 30%), MEP (19% ~ 22%) and FET (19% ~ 21%) indicators devoted high contribution to the total impact, owing to the release of $NO_X$ gas pollutants, nitrate water pollutants and heavy metal ions from power production and consumption. For these three indicators, the contribution of $H_2O_2$ is 21~36%, ~15%, 8%~10%, respectively.

### 3.2.2 US+F-AD scenario

Several studies have convinced that US-Fenton process significantly reduces the dosage of Fenton reagent under the same dewatering and oxidation efficiency (Jiang et al., 2014; Ning et al., 2014). The US-Fenton process efficiently improves the ability to dissolve SCOD in sludge

(Gong et al., 2015), thus promoting the subsequent AD efficiency. From this, the dosages of $FeSO_4$ and $H_2O_2$ set in this scenario were 66.7% and 75.0% of classic Fenton process. For the three organic contents, the total environmental impact of US+F-AD scenario was 24%, 47% and 56% lower than that of EF-AD, respectively, and the differences between the two scenarios were largely caused by FRD (28%), FEP (27%) and CC (14%). FEP (12% ~ 30%), MEP (20% ~ 21%) and FET (18% ~ 24%) also served on major contributors. The negative contribution from FRD grew obviously with the level of organic content, from -2% to -20% and -24%.

### *3.2.3 UV+F-AD scenario*

UV-Fenton process relies on the dual catalytic effect of ultraviolet light and ferrous salt to promote the decomposition of $H_2O_2$. The dosage of $FeSO_4$ and $H_2O_2$ set in this scenario was 50.0% and 33.3% of conventional Fenton process, respectively (Li, 2014). Concerning sludge with 70% and 80% organic contents, UV+F-AD scenario realized an overall environmental benefit (Table S2-8), which was inseparable from the dominant advantage of reducing FRD (-36%), carbon emissions and eutrophication level (CC: -18% ~ -19%, FEP: -14% ~ -17%). The benefits of UV+F-AD scenario obtained from avoided products for all impact categories were greater than those of EF-AD and US+F-AD for sludge with the same organic content. Whereas, these benefits did not neutralize the CHP emissions (including CO, $NO_X$, $SO_2$, NMVOCs with low ecotoxicity), which were related to MEP and acidification categories. Therefore, it is imperative to reduce the pollutant emission during CHP, optimize the heating and steam distribution mode of CHP units, and develop efficient desulfurization and denitrification agents with low environmental load (Li et al., 2019).

### *3.2.4 Comparative analysis*

The ranking of environmental impact of the three above scenarios was: EF-AD>US+F-AD>UV+F-AD (Fig. 5), owing to the maximum reduction of Fenton reagent and energy consumption by UV-Fenton process (Table S1-3). As reported in previous research, US/UV/Electro-Fenton processes increased biogas production by up to 40%~60% (Feki et al., 2020; Heng et al., 2017; Pilli et al., 2016). Whereas UV+F-AD scenario was inferior to F-AD, considering various impact indicators. The total environmental load under 60% organic content was 2.5 times of F-AD, and the total environmental benefits under 70% and 80% organic contents were reduced by 18% and 22%, for distinction in the FET (40% - 48%) and FEP (41% - 45%).

There were marginal contribution of sludge dewatering, transportation and land use process (0% ~ 1%) with ignorable environmental threats. The AD process brought significant offsetting effect for environmental load, which increased with organic content (Fig. S2-7 ~ S2-9). Regarding the contribution analysis of the key substances (Fig. S2-10), energy consumption was proved to be the decisive one (accounting for 25% ~ 90%), followed by $H_2O_2$. Diminishing energy consumption and improving energy recovery efficiency of sludge biogas are momentous for US/UV/Electro-Fenton processes.

**Fig. 5**

### *3.3 Probabilistic accuracy of environmental impact results*

Considering the significance and contrast of the environmental impact of the above different management scenarios, CC, TAP, POF, PMF and MEP were selected as observation targets, which are also prevalent among the LCA of solid waste (Bernstad and la Cour Jansen, 2012). A scientific evaluation system was constructed for result reliability, which was affected by random variability and parameter uncertainty. The identification of key process parameters and the setting of probability distribution function are shown in Section S3.

The figure of relative probability distribution function is designed to show the distribution of the impact change, while the cumulative probability distribution identified percentages corresponding to specific impact values. There was a high probability that landfill and incineration scenario suffered more from CC (Fig. 6). Meanwhile, both of F-AD and F-L scenarios were worse than the controls, but the F-I scenario was absolutely superior to the control. Based on the confidence coefficient of 95%, AD-80% scenario can avoid at least 254.11 kg $CO_2$-eq. While the cumulative probabilities for F-AD-80% and AD-70% scenarios to achieve such reduction were 76.5% and 84.6%, respectively, and for F-I-80% and Incineration-80% scenarios were zero. There was 18.8% probability that F-AD-80% would prevail over AD-70%, and the AD-80% scenario ranked first for the CC improvement, followed by AD-70% and F-AD-80%. As for MEP, PMF, POF and TAP, AD-80% scenario also showed dominant reduction under the 95% confidence interval. And the F-L and F-I scenarios showed a similar tendency as above CC (Fig. S3-2).

At the confidence interval of 95%, UV+F-AD scenario can avoid at least 31.00 kg $CO_2$-eq, while the cumulative probabilities of achieving such reduction for US+F-AD and EF-AD scenarios were 26.7% and 1.7%, respectively (Fig. 7). And the probability should be 65.3% and 50.4% for MEP; 4% and 0% for PMF; 48.9% and 26.1% for POF; 50.1% and 28.2% for TAP (Fig. S3-4). For the five impact indicators, the UV+F-AD scenario exhibited lower environmental impacts with absolute preponderance. The impact fluctuations of US+F/UV+F/EF-AD scenarios were restricted under different organic contents, bringing about significant overlap between the frequency distribution curves. High energy input are inescapable defects for the US/UV/Electro-Fenton processes, issuing in finite contribution of energy recovery and negligible improvement of environmental benefits.

**Fig. 6**

**Fig. 7**

### *3.4 Energy balance*

The comprehensive comparisons of energy balance between AD and incineration routes were presented in Fig. 8, for sludge pretreated by Fenton and US/UV/Electro-Fenton processes. The results illustrate that the energy efficiency of AD scenario (EBR: 3.3~9.0) was preferable to incineration. Adequate supply of energy produced in AD scenario not only maintains the system operation, but outputs residual energy (electricity or heat). The energy output efficiency rose sharply with the organic content. The EBR of F-AD scenarios were 42% ~ 50% higher than that of the controls, highlighting that Fenton treatment effectively optimized the anaerobic biodegradability and sludge added value. The EBR of incineration and F-I scenarios ranged from 0.2 to 0.5, denoting unattainable energy self-sufficiency. The high energy demand in management system virtually hindered EBR improvement of Fenton-pretreated sludge.

Among the US/UV/Electro-Fenton processes, EF-AD scenario presented the lowest EBR. The EBR of US+F-AD scenario was approximately 14% higher than EF-AD, while still below zero. The EBR of UV+F-AD was greater than 1, implying that the whole system could output residual energy. Noticeably, the EBR of UV+F-AD scenario was far below that of AD and F-AD, with gaps of 2.1 ~ 3.8 and 3.5 ~ 6.8, respectively. The energy potential of the UV+F-AD system is limited to the dependence on external input energy. Therefore, strengthening sludge oxidation disintegration, enhancing biogas production and reducing energy consumption are the keys to enhancing the energy balance of US/UV/Electro-Fenton processes.

**Fig. 8**

## *3.5 LCC analysis*

To comprehensively analyze the strengths and weaknesses of Fenton and US/UV/Electro-Fenton pretreatment, LCCs for seven scenarios with energy recovery were implemented (Fig. 9). The total LCCs of the three conventional scenarios ranked as: incineration > AD > landfill, in which the construction investment accounted for 90.8%, 98.6% and 94.4%, respectively, while the environmental costs accounted for 0.32%, -0.13% and 0.05%, respectively. Therefore, the differences were mainly derived from the construction costs (Table S4-3). The equipment costs of the drying beds and incinerator were relatively high, and it can be predicted from previous studies that the energy balance and economy of the sludge co-incineration or incineration ash applied for building material could still be inferior to the AD scenario (Li et al., 2017). The construction cost of the landfill scenario was lower than that of AD. There may be an underestimation of land resources, since landfill sites are closely linked with government supports, unaffected by fluctuation of the real estate market. AD scenario brought benefit income in energy recovery and environmental cost, and the benefits increased synchronously with the organics.

Turning to US/UV/Electro-Fenton process, the total LCCs ranked as: EF-AD > US+F-AD > UV+F-AD, which was consistent with the environmental impact. In view of the lack of engineering examples, the cost data of electrochemical, UV and US equipment are inaccessible, and merely covers a fraction of the overall construction cost, thus they are excluded. US+F-AD owned the lowest reagent cost, followed by UV+F-AD. The energy cost of EF-AD scenario was similar to US+F-AD, while that of UV+F-AD was just about one third of them. However, in accordance with environmental cost, only UV+F-AD and US+F-AD-60%/70% scenarios presented negative cost, especially UV+F-AD brought higher environmental benefits than F-AD. The LCC of UV+F-AD was close to that of F-AD, both of which were significantly less than their controls. The effect of Fenton and US/UV/Electro-Fenton pretreatments on LCC reduction was obvious, in which the decline increased with the organic content.

**Fig. 9**

# *4. Further Discussion*

## *4.1 Comprehensive assessment*

Overall, the total environmental impact of each sludge management scenario ranked as follows: Incineration> F-I > F-L > Landfill > EF-AD > US+F-AD > UV+F-AD >F-AD > AD. The organic contents are inversely proportional to the overall environmental impact. However, slight changes to the environmental load were recognized in those high-energy-demand technologies (e.g., incineration and US/UV/Electro-Fenton processes). As for the energy balance: F-AD> AD > UV+F-AD > US+F-AD> EF-AD> F-I > Incineration. The total LCCs of the sludge management scenarios combined with Fenton pretreatment were mostly lower than the control, ranking as: Incineration> F-I > Landfill > AD > EF-AD > US+F-AD > UV+F-AD >F-AD > F-L. LCC of F-AD scenario reached a relatively low point, slightly less than that of UV+F-AD scenario, due to the sludge reduction effect of Fenton process.

The diversity of assessment methods and localized conditions make it arduous to compare the results with other similar literatures. Climate change, freshwater eutrophication and human toxicity were widely adopted by peer experts, and played a vital role in LCA and LCC assessment. From this, they were taken as the representatives in the following comparisons. The GHG for conventional landfill scenarios in previous studies ranged from 300 to 4000 kg $CO_2$-eq/t DS (Lam et al., 2016; Xu et al., 2014), higher than the results in this research (106.2~204.6 kg $CO_2$/t DS). This difference mostly came from the electricity consumption and corresponding atmospheric emissions based on industry averages, distinct from the specific filed data in existing researches. The impacts of AD route (-310.6 ~ -66.1 kg $CO_2$/t DS) are also around the lower range of the literature values (-280 ~ 4800 kg $CO_2$/t DS), mainly because of the selection of fertilizer avoided (Lam et al., 2016; Murray et al., 2008; Xu et al., 2014). There is a similar tendency for incineration, as the results of current work (238.2~1649.2 kg $CO_2$/t DS) are within the lower range reported in the literature (130 ~5800 kg $CO_2$/t DS), considering different heat generation potential and auxiliary fuel (Houillon and Jolliet, 2005; Lam et al., 2016).

As for FEP, landfill routes in current study tends to get higher impact than the literature data ($1.2\times10^{-2}$ ~ $4.3\times10^{-2}$ kg P/t DS vs. $-6.5\times10^{-3}$ ~ 17.9 kg P/t DS), as a consequence of neglecting landfill gas recovery (Lam et al., 2016; Xu et al., 2014). The impact of AD route (-0.1~ $-2.8\times10^{-3}$ kg P/t DS) is within the range of the previously reported values (-0.1 to $-1.0\times10^{-3}$ kg P/t DS), in spite of the differences in recovery benefit of heat and power (Gourdet et al., 2017; Xu et al., 2014). Incineration routes falls in the lower range of literature data, as this study executed holistic quantification for waste heat recovery, thus decreasing phosphate release (0.2~0.4 kg P/t DS vs 0.015~18.7 kg P/t DS) (Lam et al., 2016; Tarpani et al., 2020).

From the perspective of HT, the outcomes in landfill routes (607.4~2372.8 kg 1,4-DCB/t DS) were distinct from those peer-reviewed results (8.04~830 kg 1,4-DCB/t DS) (Lam et al., 2016; Xu et al., 2014), due to the emissions of selenium and barium in the life cycle of $H_2O_2$. HT results in AD routes exceed the range of those in published papers (-3617.5~ -408.5 kg 1,4-DCB/t DS vs. -85.9~330.93 kg 1,4-DCB/t DS), in which extra electricity was applied in the further drying for digestate (Gourdet et al., 2017; Xu et al., 2014). In incineration routes, notable difference between this study and literature results was observed (9803.5~ 28645.9 kg 1,4-DCB/t DS vs. 47~1100 kg 1,4-DCB/t DS) (Lishan et al., 2018; Tarpani and Azapagic, 2018). The regional disparity of electricity, coal consumption and hazardous substances evaluation are responsible for that.

Above comparisons indicated the similar trends in the results of this study and those in previous studies, which partly supports the results validity of current study, considering the differences in assumptions, system boundaries and data.

## *4.2 Analysis of the technical barrier*

Sensitivity analysis was adopted to optimize the F-AD scenario. Considering the optimal performance of F-AD scenario under 80% organic content and key process parameters mentioned above, the sensitivity analysis was conducted by selecting two pivotal factors: the biogas heat recovery efficiency in AD process and the environmental impact reduction of $H_2O_2$ upstream production. The two factors were altered in selected rangeability, and the differences between the total impact potentials of F-AD-80% and AD-80% scenarios were calculated. The contour line shown in Fig. 10 visualizes the dynamic changes of scenario optimization under the synergy of the two factors. The blue area above the line marked 0.0 shows the parameter variation range when the impact potential of F-AD-80% is lower than AD-80%, that is, F-AD scenario will be preferred than AD within this range. The total impact potential tended to dramatically fluctuate, and the environmental impact variations involved in $H_2O_2$ took a stronger role in the results. For the optimization of F-AD scenario, it is highly crucial to simultaneously fulfill a technical breakthrough in biogas recovery and $H_2O_2$ production industry.

**Fig. 10**

### *4.3 Outlooks*

Visibly, the incorporation of Fenton process was deemed to be environmentally unfriendly based on the above results of LCA. Mitigation strategies such as exploring optimization technologies to minimize the amount of $H_2O_2$ and waste energy or developing alternative chemicals for $H_2O_2$ (e.g., $O_3$, ferrate) should be the hotspots. Further research ought to be implemented to achieve effective utilization of UV radiation in sludge mixture with high solid content. By extension, the potential of natural daylight in Fenton process may also serve as a promising strategy to clear the obstacles of environmental hazard for photo-Fenton application.

Ultimately, some scarcities in this research need pointing out, which should be addressed in future studies. Firstly, more accurate engineering data is required to facilitate in-depth research. The optimal dosages of Fenton reagents and process conditions selected in this study may be inapplicable to the sludge with special properties, expecting for more well-designed simulations. There might be a transfer of environmental burden from sludge to waste liquid with increased iron content after sludge dewatering. Therefore, a system boundary expansion to cover the subsequent wastewater treatment process is a major issue of future study. Simultaneously, LCA associated with other AOP systems for sludge is conducive to the horizontals and vertical comparison between different oxidation processes.

## *5. Conclusion*

In this paper, with the aid of LCA and LCC, the environmental impact caused by the incorporation of Fenton processes into sludge management scenarios were systematically analyzed, and the essential processes and substances were identified. For the selected scenarios, sludge organic content has slight influence on the total environmental impacts (excluding F-AD scenario), as well as economic performance, but has significant influence on their energy efficiencies. Considering environmentally friendliness, energy efficiency and economic effect, F-AD scenario seemed to be the reasonable trade-off, based on the assumptions and collected data. Strong evidence was also provided for the prospect of collaborative F-AD system for urban multi-source wastes with high organics (household garbage, waste oil, food residue, etc.). Fenton process is warmly welcomed for its advantages in sludge reduction, deodorization, disinfection, and degradation of toxic substances. Whereas, this study recognized that Fenton systems potentially provide undesirable threats to natural environment (FDP, FEP, MEP, CC, etc.). When it is inaccessible to improve $H_2O_2$ upstream production process, we should focus on searching its substitutes and low-energy processes with superior oxidation capability (e.g., $O_3$) and environmental friendliness in the future.

## *Credit authorship contribution statement*

**Rourou Zhang:** Methodology, Data curation, Data analyses. **Xiao Liu:** Data analyses, Writing assistance. **Renglu Chen:** Data analyses. **Zijing Wang:** Writing assistance. **Wei Lin:** Data analyses, Writing assistance. **Huu Hao Ngo:** Conceptualization, Methodology, Editing. **Jun Nan:** Conceptualization, Methodology, Editing. **Guibai Li:** Conceptualization, Methodology, Supervision. **Jun Ma:** Conceptualization, Methodology, Editing. **An Ding:** Conceptualization, Methodology, Editing, Supervision.

## *Declaration of competing interest*

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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# Figure Captions

**Fig. 1** System boundary........................................................................................................................ 1

**Fig. 2** Comparison of environmental impact relative results in sludge landfill route. For each impact indicator, the larger value in the two scenarios was set to ±100%, and the smaller value of the other scenario was shown as a percentage relative to the larger one............................................................... 2

**Fig. 3** Comparison of environmental impact relative results in F-AD route.......................................... 3

**Fig. 4** Comparison of environmental impact relative results in F-I route. ............................................. 4

**Fig. 5** Comparison of environmental impact relative results in US+F/UV+F/EF-AD route. Organic contents: (a) 60%, (b) 70%, (c) 80%. .......................................................................................... 5

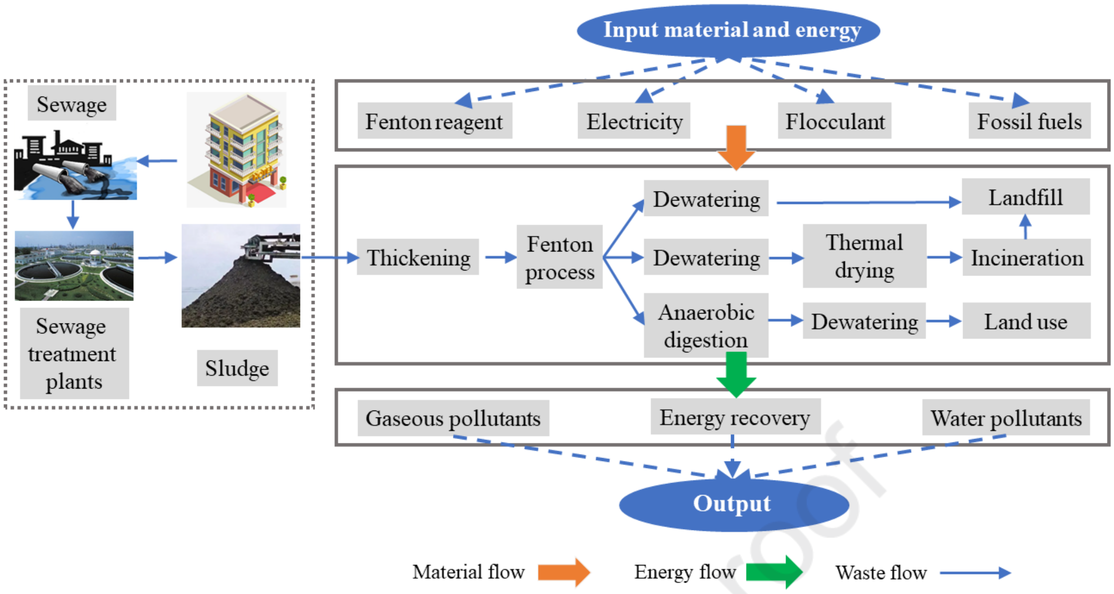
**Fig. 6** Cumulative frequency distribution of climate change for F-L, F-AD, F-I and their control scenarios. ........................................................................................................................ 6

**Fig. 7** Cumulative frequency distribution of CC for US+F/UV+F/EF-AD scenarios............................. 7

**Fig. 8** EBR of sludge treatment and disposal scenarios with different organic contents. ....................... 8

**Fig. 9** LCCs of seven sludge treatment and disposal options. (Sludge with three levels of organic contents based on AD and incineration treatment).................................................................................. 9

**Fig. 10** Contour lines of the differences between the total impact of F-AD-80% and AD-80% scenarios. ........................................................................................................................ 10



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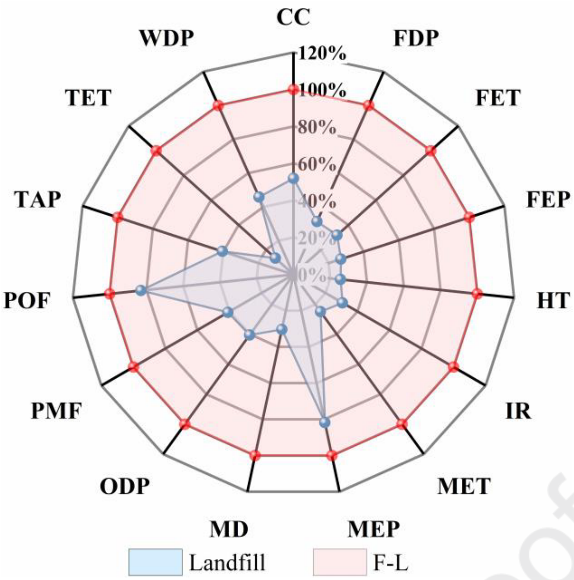


**Fig. 2** Comparison of environmental impact relative results in sludge landfill route. For each impact indicator, the larger value in the two scenarios was set to ±100%, and the smaller value of the other scenario was shown as a percentage relative to the larger one.

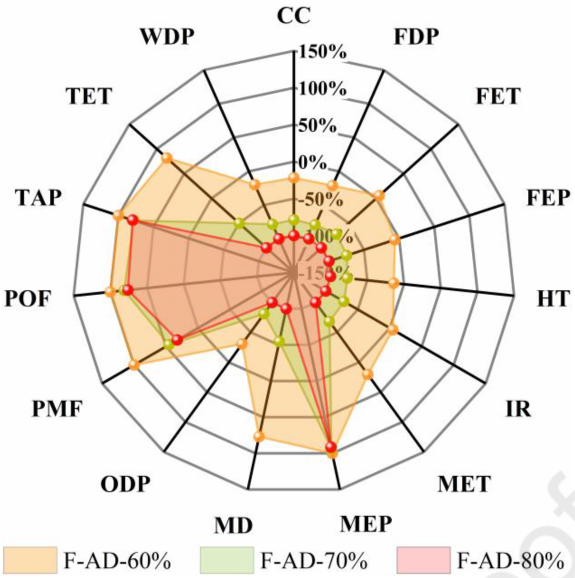


**Fig. 3** Comparison of environmental impact relative results in F-AD route.

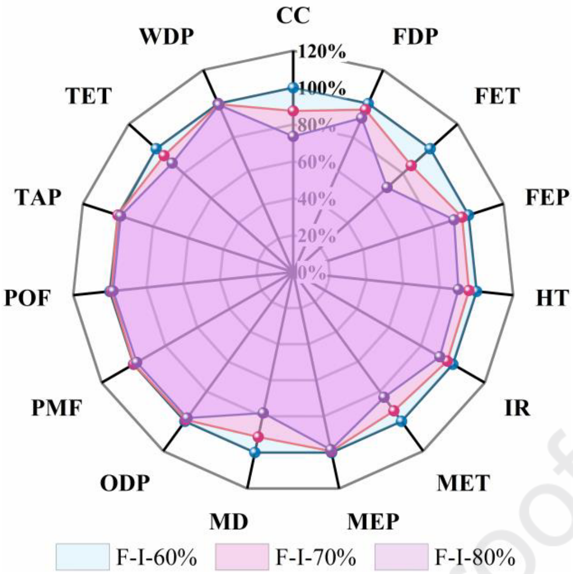
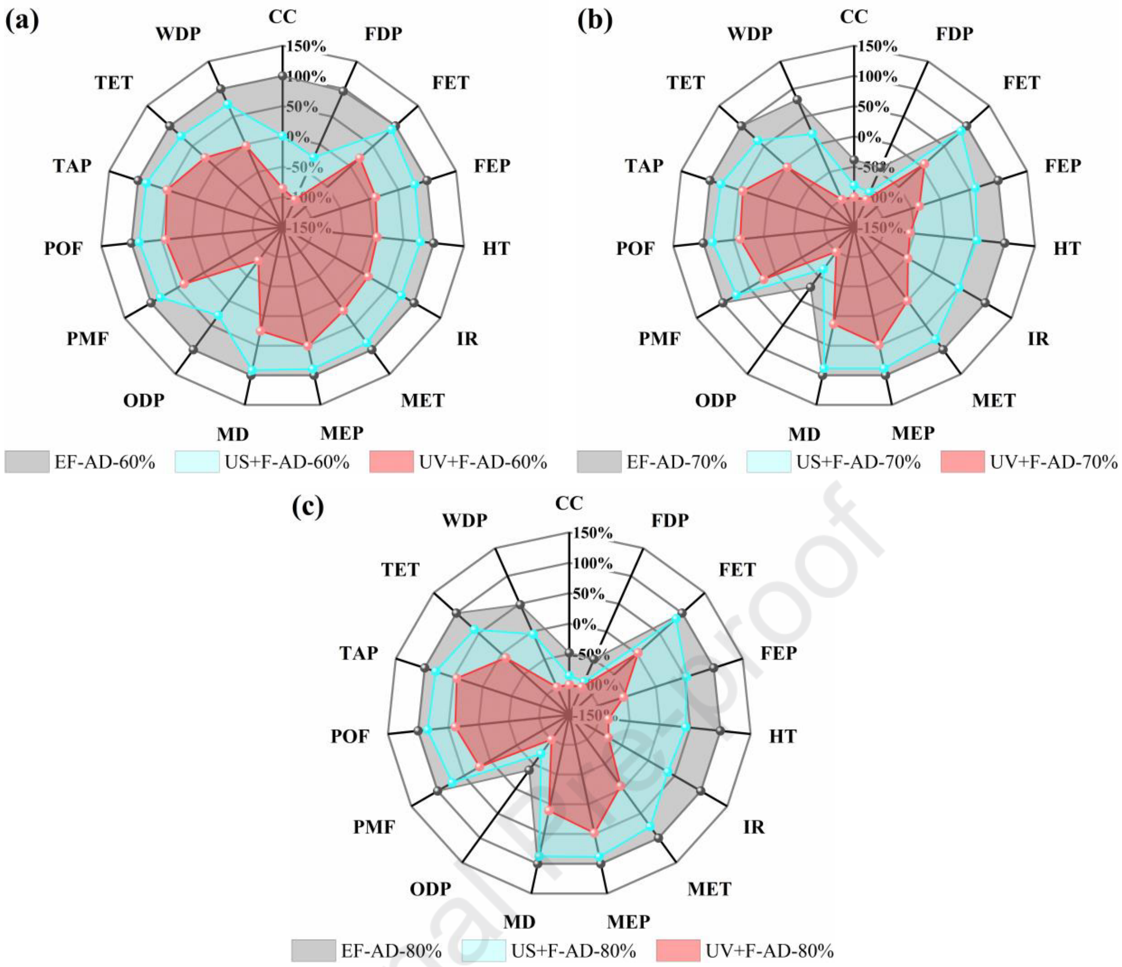


**Fig. 4** Comparison of environmental impact relative results in F-I route.



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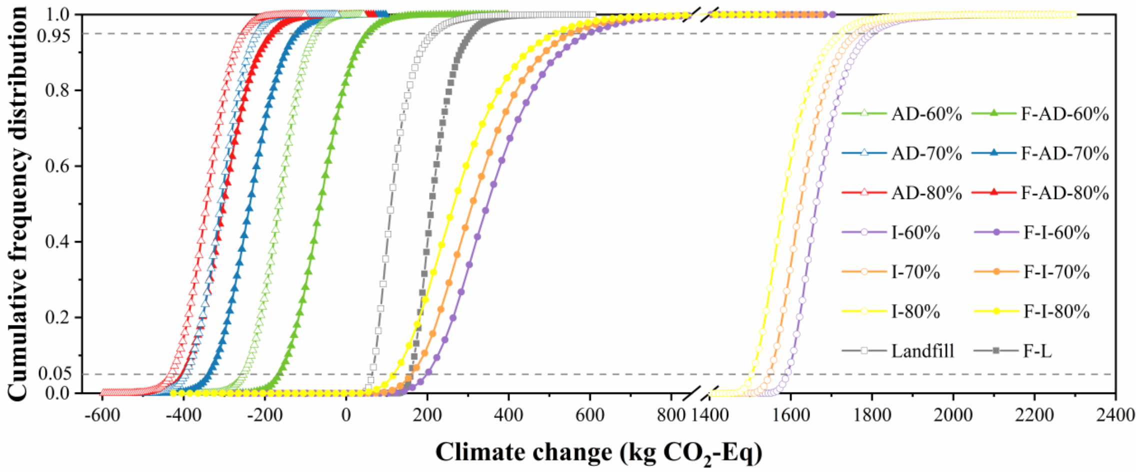
Organic contents: (a) 60%, (b) 70%, (c) 80%.



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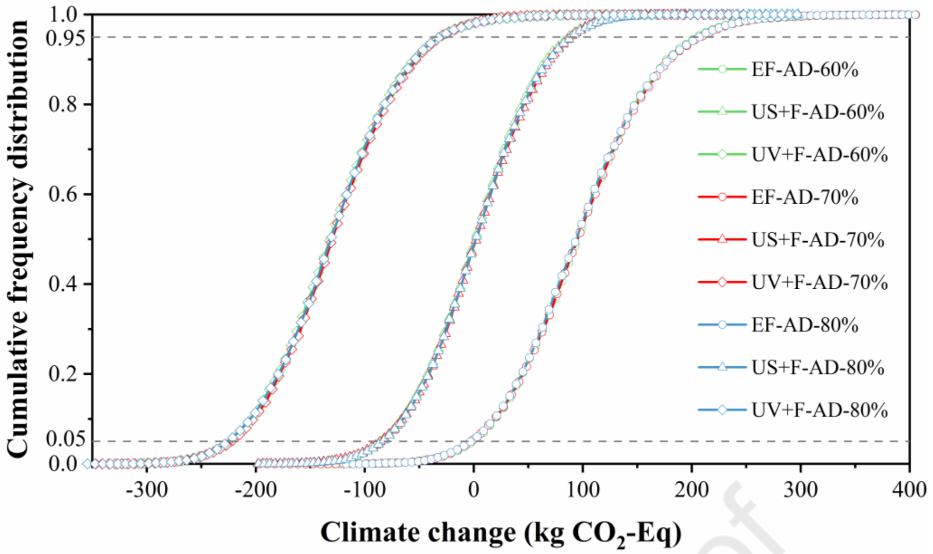
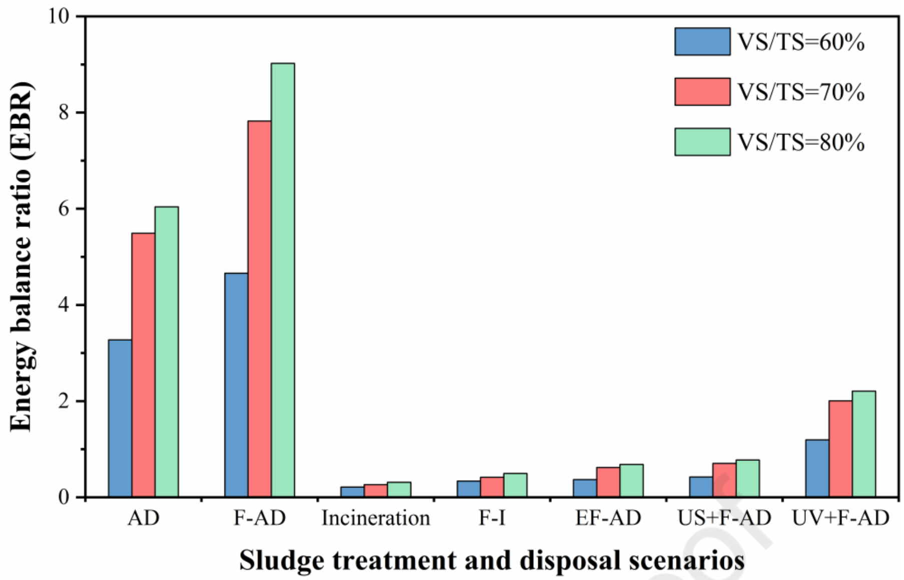


**Fig. 7** Cumulative frequency distribution of CC for US+F/UV+F/EF-AD scenarios.



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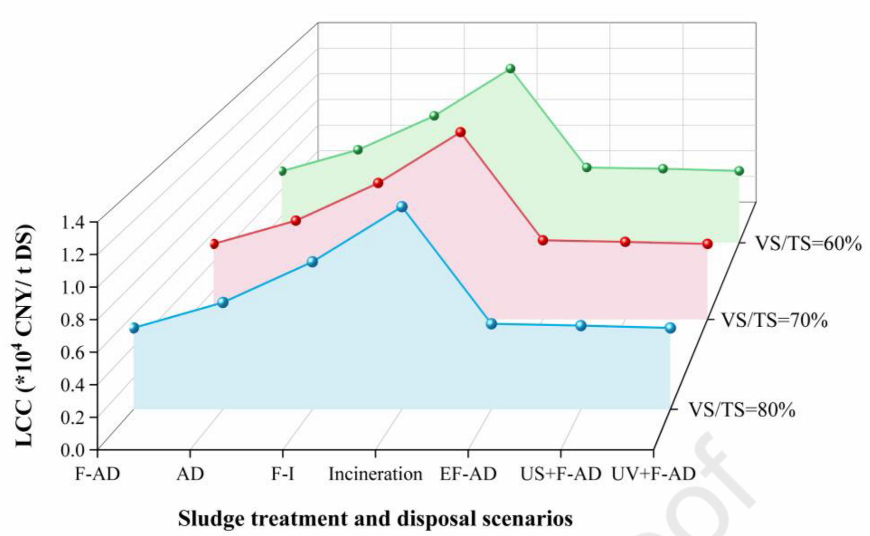

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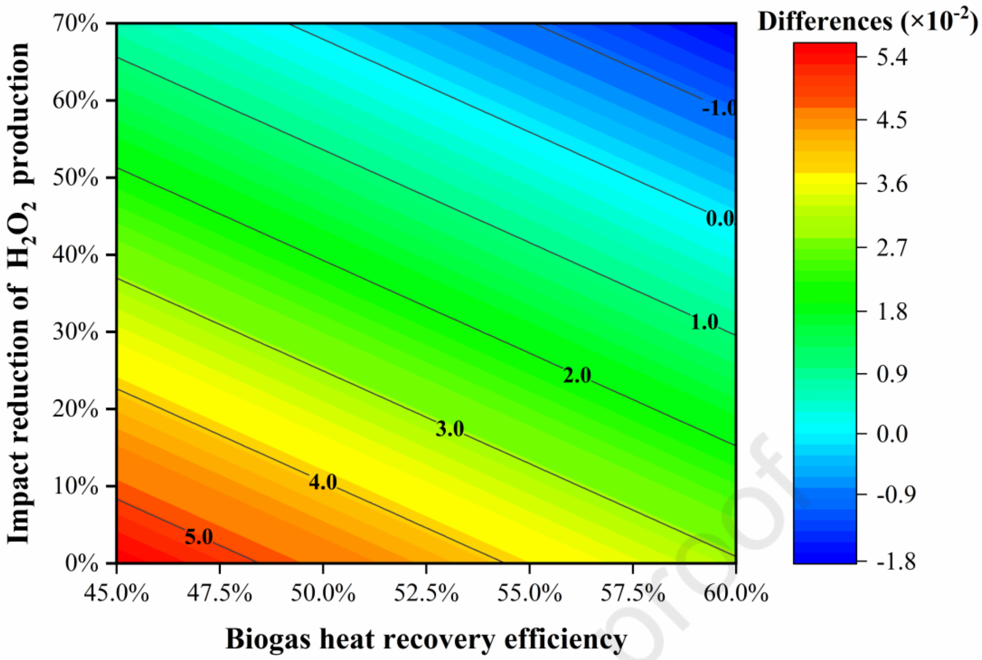

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