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Environmental impact of the effluents discharging from full-scale wastewater treatment plants evaluated by a hybrid fuzzy approach

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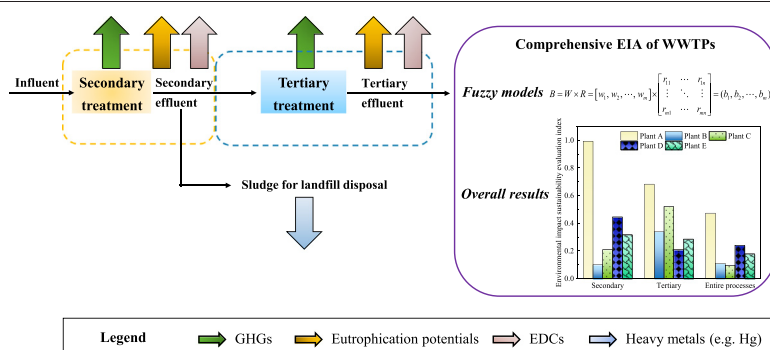
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HIGHLIGHTS

- Comprehensive environmental impact analyses of five full-scale WWTPs are studied.
- Biological process and electricity use contribute largely to GHGs during treatment.
- Special concerns need to be paid to EDCs such as NP1EO and E1 during treatment.
- The potential ecological risks of heavy metals in sludge (e.g. Hg) were identified.
- Plant A achieved highest performance in comprehensive EIAs among all WWTPs.

GRAPHICAL ABSTRACT



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ABSTRACT

Increasing attention is being paid to the environmental impacts of wastewater treatment plant (WWTP) effluent. In this study, comprehensive environmental impact analyses (EIAs) were performed for the secondary treatment processes, tertiary treatment processes, and entire plants at five full-scale WWTPs in Kunming, China. The EIAs took into account greenhouse gas (GHG) emissions, potential for the effluent to cause eutrophication, ecological risks posed by endocrine disrupting compounds (EDCs) in treated effluent, and the risks posed by heavy metals in excess sludge. A comprehensive assessment toward environmental sustainability was performed using a fuzzy approach. The results indicated that the biological treatment process made the largest contribution (>68% of the total) of the secondary treatment processes to GHG emissions and that electricity consumption made the largest contribution (>64% of the total) of the tertiary treatment processes to GHG emissions. Large numbers of EDCs were removed during the secondary treatment processes, but the potential ecological risks posed by EDCs still require attention. High mercury concentrations were found in excess sludge. The plant that removed the largest proportion of pollutants and produced effluent posing the least ecological risks gave the best comprehensive EIA performance.

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1. Introduction

Treating wastewater is vital to protect human health and the environment. Most centralized wastewater treatment plants (WWTPs) currently use a combination of physical, chemical, and biological treatment processes to remove pollutants (e.g., organic matter, suspended solids, nutrients, and pathogens) from wastewater (Chen et al., 2018). However, large amounts of energy and materials are used, pollutants are discharged, and byproducts are generated during wastewater treatment processes (Molinos-Senante et al., 2014b; Wang et al., 2018). For example, large amounts of carbon dioxide (CO₂) are generated during aerobic oxidation, which is the main WWTP process that emits greenhouse gases (GHGs) (Nguyen et al., 2020). Methane (CH₄) and nitrous oxide (N₂O), which are GHGs that can have strong effects, can be released to the atmosphere under anoxic and/or anaerobic conditions (Lorenzo-Toja et al., 2016; Chen et al., 2017). Nutrient removal techniques are used in many WWTPs, but some nutrients (e.g., nitrogen and phosphorus), organic matter, and emerging pollutants (e.g., trace organic compounds and antibiotic-resistant bacteria) can still be present in the sludge produced. Other nutrients can enter water bodies receiving treated effluent. These nutrients can cause eutrophication, can be toxic, and can pose other risks to the environment (Khiewwijit et al., 2018; Awad et al., 2019).

New concepts and management models for sustainable wastewater treatment have been proposed to address the challenges described above. It has been claimed that effort should be made to intensify processes, recover resources, and integrate systems to improve effluent quality and to achieve zero discharge and neutral carbon emissions (Chhipi-Shrestha et al., 2017; Soares, 2020). In particular, it has been stated in many publications that wastewater utilization should be improved by promoting water reuse, recovering nutrients, metals, and biomaterials, and transforming organic carbon into biogas (Khiewwijit et al., 2018; Qu et al., 2019). Some studies have been focused on optimizing systems, integrating water supply, wastewater drainage, wastewater treatment, and wastewater reclamation, and matching urban water systems to the socioeconomic situation (Lane et al., 2015; Su et al., 2019). It is important to evaluate and minimize environmental impacts during wastewater treatment and to assess the risks posed to the environment receiving the effluent and sludge produced, to move toward the development of sustainable WWTPs.

Many studies of the environmental impacts of WWTPs have been performed (Stokes and Horvath, 2010; Corbella et al., 2017; Kamble et al., 2018). It has been found that the environmental impacts of WWTPs are mainly associated with operating WWTPs and that constructing and demolishing WWTPs contribute relatively little to the total environmental impacts (Rodriguez-Garcia et al., 2011; Awad et al., 2019). Gaseous emissions and energy consumption are the main contributors to the environmental impacts of WWTPs (Chen et al., 2018; Awad et al., 2019). The main efforts to quantify direct GHG emissions from WWTPs have involved monitoring CO₂, CH₄, and N₂O emissions during wastewater treatment (Lorenzo-Toja et al., 2016). In some studies it was found that N₂O was emitted from anoxic zones and pre-sedimentation tanks, secondary clarifiers, nitrification zones, and sludge treatment units (Czepiel et al., 1995). It is therefore important to gain an understanding of the environmental impacts of typical treatment processes and the entire treatment procedures used in WWTPs.

The life cycle assessment (LCA) method, a type of environmental impact assessment (EIA), has been used in some studies to assess the environmental impacts of WWTPs. Various impacts, such as GHG emissions, eutrophication, acidification, photochemical oxidation, ecotoxicity, ozone layer depletion, and abiotic resource depletion, can be taken into account in an EIA. However, life cycle inventory databases do not contain all the data required for an EIA, so national mean values and data from various publications are used when performing EIAs. This

may lead to large differences between EIA results and actual environmental impacts caused by operating WWTPs (Chen et al., 2012; Corominas et al., 2013). Various life cycle impact assessment (LCIA) approaches are available (e.g., CML, ILCD, TRACI, EDIP 2013, Eco-indicator 99, IMPACT 2002+, and ReCiPe), and different methods may give different results for some impact categories (Wittmaier et al., 2009). LCAs have therefore often been used to assess differences between different treatment techniques and management practices (Kamble et al., 2018) rather than to attempt to accurately quantify the environmental impacts of WWTPs (Corominas et al., 2020). GHG emissions and eutrophication potentials are the dominant contributors to the environmental impacts of WWTPs (Rodriguez-Garcia et al., 2011; Lorenzo-Toja et al., 2016; Dong et al., 2017; Chen et al., 2018; Wang et al., 2018). Therefore, for simplicity, EIAs of WWTPs mainly focused on GHG emissions and eutrophication potentials, and the other impact categories are often neglected.

Rapid increases in the numbers of WWTPs in urban areas, particularly in developing countries, in recent years have led to increasing attention being paid to the risks posed to human and environmental health by contaminants discharged from WWTPs and the waste produced by WWTPs (Rashid and Liu, 2021). Methods for determining the effects of different wastewater treatment processes on human health and the environment using biological assays and conversion models for performing overall assessments have recently been developed (Papa et al., 2016; Pedrazzani et al., 2019). The risks posed to the environment by emerging pollutants (e.g., pharmaceuticals and personal care products (PPCPs) and endocrine disrupting compounds (EDCs)) in WWTP effluent and by heavy metals in sludge produced in WWTPs have been evaluated (Mohapatra et al., 2016; Tytla, 2019; Zhou et al., 2019). However, the risks posed by contaminants have been incorporated into EIAs of WWTPs to achieve comprehensive evaluations in few studies. LCAs of WWTPs cannot directly provide detailed information on the risks posed to the environment (Corominas et al., 2020). Large differences have been found between the toxicity impacts calculated using different LCIA approaches, and some possible additive, synergistic, or antagonistic effects of different substances are not considered when performing conventional assessments (Pedrazzani et al., 2019). Characterization factors and impact scores for specific PPCPs, EDCs, and heavy metals still need to be investigated (Zang et al., 2015; Li et al., 2019). Overall, local investigation is preferred over computing or modeling assessments for trace organic pollutants and heavy metals because trace organic pollutant and heavy metal concentrations and behaviors may be very different for different treatment techniques, at different locations, and in different seasons (Rashid and Liu, 2021).

The present study aims to conduct comprehensive EIAs of the secondary, tertiary and entire processes in several full-scale WWTPs, to determine GHG emissions, eutrophication potentials and ecological risks. Decision making is complicated and often involves dealing with uncertainties. A simple assessment method using a set of indicators cannot give an holistic assessment because the value of each indicator will be related separately to each aspect of the system being assessed (Molinos-Senante et al., 2014a). Appropriate aggregation techniques are therefore required to derive composite indicators to allow multi-dimensional assessments and comparisons to be performed. Multiple attribute decision making (MADM) techniques (e.g., the analytic hierarchy process, data envelopment analysis, and the technique for order preference by similarity to an ideal solution) have therefore been developed and are being used increasingly often. However, in many MADM studies, the factor weightings are often chosen subjectively, which may lead to unscientifically and irrationally justified results (Chen et al., 2014). Therefore, a hybrid fuzzy approach was used in this study, using membership functions that reflected various factors well and ensured that the evaluation was objective and rational (Tan et al., 2014). The results are expected to help wastewater treatment processes to improve WWTP sustainability at the local, regional, and even global scales.

2. Methods

2.1. Overview of the WWTPs and treatment processes

Five WWTPs in the central parts of Kunming City in Yunnan Province, China, were used in the study. The overall capacities of the WWTPs and the treatment processes used in the WWTPs are shown in Table S1. The operating parameters for the WWTPs are shown in Table S2. Each plant used three treatment levels (primary, secondary, and tertiary treatments) to ensure that the effluent met stringent discharge standards. Dewatered sludge produced in each plant was transported away from the plant for disposal. Plant A used an oxidation ditch process (i.e., Carrousel and Orbal processes in phases 1 and 2, respectively) as a secondary treatment. Plants B and D both used anaerobic–anoxic–oxic (A²O) processes as secondary treatments. Plant C used the intermittent cycle extended aeration process (ICEAS) with continuous inflow/intermittent outflow and no internal/external reflux as a secondary treatment. Plant E used the University of Cape Town (UCT) process (in which the sludge from the settling tank is transferred into the anoxic tank rather than the anaerobic tank) as a secondary process.

All five WWTPs used tertiary treatments such as coagulation/flocculation, filtration, and disinfection to remove total suspended solids (TSS), total phosphorus (TP), and pathogens. Plant A did not contain a coagulation/flocculation tank, and the secondary treatment effluent was fed directly into a D-type filter tank (Dean Corporation, China). The A²O process used in Plant B was performed using an anaerobic pond and a concentric cycle degradation pond in which the inner circle of the concentric circle was the anoxic zone and the outer ring was the aerobic zone. Plant B did not contain a coagulation/flocculation tank, but the long distance between the secondary sedimentation tank and tertiary filtration tank allowed space for the coagulant to be added and for the coagulant to mix and react with the wastewater. The effluent was then fed into a V-type filter tank (Degrémont, France). Plant C contained an ACTIFLO system, which contained a coagulation basin (in which polyaluminum chloride (PAC) was used as a coagulant), a dosing basin (in which polyacrylamide (PAM) and microsand were added), a flocculation basin (in which PAM was used as a flocculant), and a sedimentation basin. Plant C also contained a D-type filter tank (Dean Corporation, China). Plant D contained a coagulation basin and a V-type filter tank (Degrémont, France). Plant E contained a folded-plate flocculation basin and a D-type filter tank (Dean Corporation, China). The D-type filters used fibers rather than quartz sand as the filtration medium. The fiber filters were small but highly efficient.

Samples of the influents and effluents from the secondary and tertiary process systems in each plant were collected once each day during the experiment between 09:00 and 11:00. Each sample was stored in a sterile glass bottle shielded from the light. The samples were transported to the laboratory in ice-packed coolers and were analyzed

within 4 h after collection. The water quality parameters were determined using standard methods for analyzing water and wastewater (APHA, 2017). The water quality data of interest for all five plants were collated each week for one-year period and then the GHG emissions and eutrophication potentials for the plants were calculated. Data for nine EDCs in effluent and eight heavy metals in sludge were taken from publications by Huang et al. (2014) and Li et al. (2015), respectively. The data were processed using Microsoft Office Pro 2016 software (Microsoft, USA) and OriginPro 9.0 software (OriginLab, USA).

2.2. System boundaries and evaluation indexes

The system boundaries of the primary processes, secondary processes, tertiary processes, and the entire treatment procedures in the five WWTPs that were used in the EIAs are shown in Fig. 1. The system boundary for an entire WWTP consisted of the primary, secondary, tertiary, and sludge handling processes. The primary treatments played limited roles compared with the biological treatments in removing organic compounds and pathogens. However, the influent data were collected from the primary treatment inflows, so the system boundaries for the secondary processes incorporated both the primary and biological treatments to simplify the calculations. The system boundaries for the tertiary processes included coagulation/flocculation, filtration, and UV disinfection. The influent and effluent data for the tertiary processes were the secondary treatment effluent and UV disinfection effluent, respectively. It is essential to use appropriate evaluation indexes when performing an EIA. As shown in Table 1, the EIA for each WWTP was focused on GHG emissions, the eutrophication potentials, and the ecological risks.

2.3. EIA calculation methodologies

The functional unit used in the study was the equivalent of removing 1 t of chemical oxygen demand (1 t COD_{eq}) from water. As suggested by Copp et al. (2002) and Benedetti et al. (2008), the COD_{eq} values for TSS, total nitrogen (TN) and TP were 2 kg COD_{eq}/kg TSS, 20 kg COD_{eq}/kg TN, and 100 kg COD_{eq}/kg TP, respectively.

2.3.1. EIA calculation of different environmental aspects

The revised Bridle model was used to calculate GHG emissions. The model and the parameters used are shown in Table S3. GHG emissions during the secondary processes occurred through the biological treatments (i.e., CO₂ generated through oxidation, endogenous respiration, CO₂ consumption during nitrification, and N₂O and methane released under anoxic/anaerobic conditions) and through electricity consumption. GHG emissions during the tertiary processes occurred through electricity and chemical consumption. PAC was the main chemical used in all five WWTPs, so GHG emissions caused by the use of polyacrylamide and microsand in Plant C were excluded from the

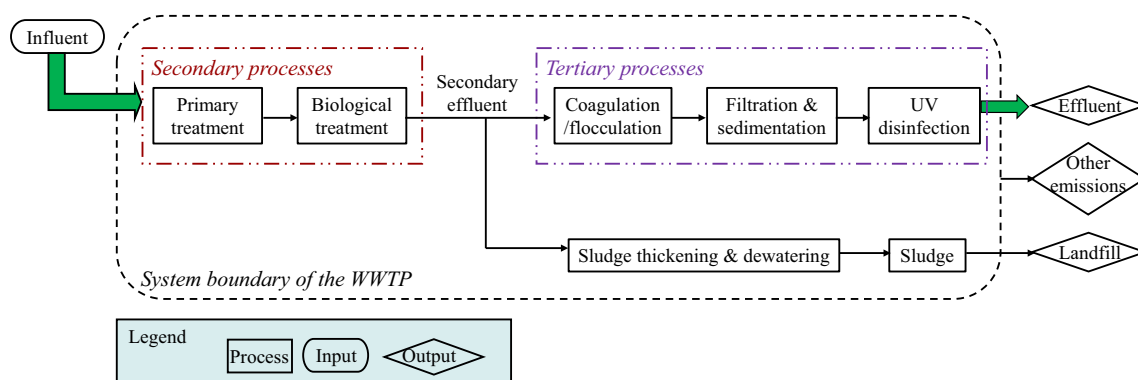


Fig. 1. System boundary of the secondary, tertiary and entire processes in EIA.

Table 1
Evaluation indexes considered for EIA under different system boundaries.

EIA indexes	Secondary processes	Tertiary processes	Entire processes of WWTP
GHG emissions	✓	✓	✓
Eutrophication potentials	✓	✓	✓
Ecological risks of EDCs	✓	✓	✓
Ecological risks of heavy metals in sludge	-	-	✓

calculations. GHG emissions for the entire treatment processes mainly occurred through the biological treatments, electricity consumption, and chemical consumption during the tertiary treatments and sludge treatments. The global warming potentials of the GHGs emitted were quantified in terms of CO₂ equivalents (CO_{2eq}) using Eq. (1). As suggested in the Intergovernmental Panel on Climate Change (IPCC) Guidelines for GHG inventories, the global warming potentials of CO₂, N₂O, and CH₄ were 1, 296, and 23, respectively (IPCC, 2006).

$$W_{CO_2eq} = (W_{CO_2} + 296 \times W_{N_2O} + 23 \times W_{CH_4}) / \Delta COD_{eq} \quad (1)$$

where,

- W_{CO_{2eq}} is the equivalent to CO₂ emissions (kg CO_{2eq}/t COD_{eq});
- W_{CO₂} is the amount of CO₂ emissions (kg CO₂/d);
- W_{N₂O} is the amount of N₂O emissions (kg N₂O/d);
- W_{CH₄} is the amount of CH₄ emissions (kg CH₄/d);
- ΔCOD_{eq} is the equivalent to COD removal amount (t COD_{eq}/d).

The eutrophication potential was defined as the degree of eutrophication caused by discharge of a pollutant per unit mass. The eutrophication potentials were expressed as PO₄-P equivalents (PO₄-P_{eq}) and calculated using Eq. (2). The eutrophication potentials for the water quality parameters TP, ammonium nitrogen (NH₄-N), nitrate nitrogen (NO₃-N) and COD were 3.06 kg PO₄-P_{eq}/kg TP, 0.33 kg PO₄-P_{eq}/kg NH₄-N, 0.1 kg PO₄-P_{eq}/kg NO₃-N, and 0.022 kg PO₄-P_{eq}/kg COD, respectively (Guinee, 2002).

$$Eutrophication_i = (EP_i \times M_i) / \Delta COD_{eq} \quad (2)$$

where,

- Eutrophication_i represents the degree of eutrophication caused by the discharge of the *i*th pollutant in the effluent (kg PO₄-P_{eq}/t COD_{eq});
- EP_i represents the eutrophication potential of the *i*th pollutant (kg PO₄-P_{eq}/kg);
- M_i represents the average daily discharge of the *i*th pollutant (kg/d);
- ΔCOD_{eq} represents the average daily removal of COD equivalent (t COD_{eq}/d).

The ecological risks posed by EDCs were assessed using data for nine phenols and steroids, including the 4-t-octyl phenol (4-t-OP), 4-nonylphenol (4-NP), bisphenol A (BPA), nonylphenol monoxyethylene ether (NP1EO), nonylphenol dioxyethylene ether (NP2EO), estrone (E1), estradiol (E2), ethinyl estradiol (EE2) and estradiol (E3). The risk quotient method was used to quantify the ecological risks using Eq. (3).

$$RQ = MEC / PNEC \quad (3)$$

where,

- RQ is the risk quotient;
- MEC is the measured concentration of the EDC (ng/L);
- PNEC is the predicted non-effect concentration of the EDC (ng/L).

It has previously been found that the PNEC values of 4-t-OP, 4-NP, BPA, NP1EO, NP2EO, E1, E2, EE2 and E3 were 120, 330, 1000, 110, 110, 6, 2, 0.1 and 60 ng/L, respectively (Fenner et al., 2002; Stasinakis et al., 2008; Chen et al., 2009; Caldwell et al., 2012). A RQ greater than 1 indicates that the EDCs are likely to pose high ecological risks. The total risk quotient was defined as the sum of the RQs for the different EDCs (Escher et al., 2011).

The potential ecological risk index was used to evaluate the ecological risks posed by heavy metals in dewatered sludge. This method is often used to evaluate heavy metal contamination of soil and sediment. The sludge was transported away from the WWTPs for further disposal, so background heavy metal concentrations in sludge were compared with the concentrations of heavy metals in soil from Yunnan Province (Table S4). The potential ecological risk index was calculated using Eq. (4). The potential ecological risk evaluation classes are shown in Table S5 (Fernández and Carballeira, 2001; Ma and Han, 2019).

$$RI = \sum_{i=1}^n E_r^i = \sum_{i=1}^n T_r^i \times C_f^i = \sum_{i=1}^n T_r^i \times \frac{C_s^i}{C_n^i} \quad (4)$$

where,

- RI represents the potential ecological risk index of various heavy metals;
- E_rⁱ is the potential ecological risk index of the *i*th heavy metal;
- T_rⁱ is the toxicity response coefficient of heavy metal *i*;
- C_fⁱ is the pollution coefficient of the relative parameter ratio of heavy metal *i*;
- C_sⁱ is the measured value of heavy metal *i* (mg/kg);
- C_nⁱ is the background reference value (mg/kg) of heavy metal *i*.

2.3.2. Fuzzy comprehensive evaluation

The fuzzy comprehensive evaluation was performed using the environmental assessment factor set *U* for the different treatment processes shown in Table 1. The evaluation sets *V* for the different treatment processes were based on the system boundaries shown in Table S1 and Fig. 1. The main steps in the evaluation were identifying the membership degree, constructing a single factor evaluation matrix, determining the weightings of the evaluation factors, and conducting the comprehensive evaluation.

For *m* evaluation indexes and *n* evaluation objects, the initial matrix was formed by Eq. (5).

$$X = (x_{ij})_{m \times n} = \begin{bmatrix} x_{11} & \cdots & x_{1n} \\ \vdots & \ddots & \vdots \\ x_{m1} & \cdots & x_{mn} \end{bmatrix} \quad (5)$$

where *x*_{*ij*} is the *i*th evaluation index corresponding to the *j*th evaluation object.

For revenue-based indexes, specifically those that the larger the attribute value is, the better the index is. The standardized index can be calculated as Eq. (6).

$$a_{ij} = \frac{x_{ij}}{x_{ij}^{\max}}, i = 1, 2, \dots, m; j = 1, 2, \dots, n \quad (6)$$

where *x*_{*i*}^{max} refers to the maximum value of the *i*th evaluation index.

For cost-based indexes, specifically, those that the smaller the attribute value is, the better the index is. The standardized index can be calculated as Eq. (7).

$$a_{ij} = \frac{x_{ij}^{\min}}{x_{ij}}, i = 1, 2, \dots, m; j = 1, 2, \dots, n \quad (7)$$

where *x*_{*i*}^{min} refers to the minimum value of the *i*th evaluation index.

The membership function was essentially a black box model. The membership function characterized the membership degree for each

evaluation index using well-established functions. The membership function therefore objectively reflected each index being a member of an evaluation grade (Chen et al., 2014; Tan et al., 2014). Determining the membership functions is a crucial part of a fuzzy comprehensive evaluation. The membership function allowed the fuzzy evaluation grade to be numerically concretized as the membership degree to allow a fuzzy set to be constructed. The membership function can be affected by the evaluation targets, data characteristics, expert experience and other factors (Chen et al., 2014). Commonly used methods for determining membership functions include fuzzy statistical analysis, fuzzy inference, binary comparison sorting, fuzzy distribution, and expert determination. The binary comparison sorting approach was used in this study.

The membership functions were divided into two types according to the index type. For a cost-based index, the minimum value of the *i*th index was used as the benchmark for comparison and the membership function is shown in Eq. (8).

$$y = \begin{cases} e^{-\left(\frac{x_{ij}}{x_i^{\min}} - 1\right)} & x_{ij} \neq x_i^{\min} \\ 1 & x_{ij} = x_i^{\min} \end{cases} \quad i = 1, 2, \dots, m; j = 1, 2, \dots, n \quad (8)$$

For a revenue-based index, the maximum value of the *i*th index was used as the benchmark for comparison and the membership function is shown in Eq. (9).

$$y = \begin{cases} e^{-\left(\frac{x_i^{\max}}{x_{ij}} - 1\right)} & x_{ij} \neq x_i^{\max} \\ 1 & x_{ij} = x_i^{\max} \end{cases} \quad i = 1, 2, \dots, m; j = 1, 2, \dots, n \quad (9)$$

The membership degree for each index was determined from the membership function, and a single-factor fuzzy evaluation matrix was constructed as shown in Eq. (10).

$$R = (r_{ij})_{m \times n} = \begin{bmatrix} r_{11} & \dots & r_{1n} \\ \vdots & \ddots & \vdots \\ r_{m1} & \dots & r_{mn} \end{bmatrix} \quad (10)$$

where r_{ij} represents the membership degree of the *i*th evaluation index of the *j*th evaluation object.

The quotient weight method was used to determine the weighting of each environmental impact index. The method used actual data and avoided bias caused by the experience of the researcher. A high degree of variation in the evaluation index will generally be more informative than a low degree of variation, so a larger weighting can be given to an index if necessary (Sun et al., 2017). The specific weighting of an index was calculated using Eq. (11).

$$f_{ij} = \frac{a_{ij}}{\sum_{j=1}^n a_{ij}} \quad i = 1, 2, \dots, m; j = 1, 2, \dots, n \quad (11)$$

where a_{ij} is the standardized value of the *i*th index of the *j*th evaluation object. f_{ij} is the specific weight of the index value.

The quotient value for the quotient weighting was calculated using Eqs. (12)–(14).

$$e_i = -k \sum_{j=1}^n f_{ij} \ln f_{ij} \quad i = 1, 2, \dots, m \quad (12)$$

$$k = \frac{1}{\ln n}, 0 \leq e_i \leq 1 \quad (13)$$

$$w_i = \frac{1 - e_i}{\sum_{i=1}^m (1 - e_i)}, \sum_{i=1}^m w_i = 1 \quad (14)$$

Table 2
Weightings of environmental impact indexes under different system boundaries.

EIA indexes	Secondary processes	Tertiary processes	Entire processes of WWTP
GHG emissions	0.0791	0.2178	0.0773
Eutrophication potentials	0.3859	0.1634	0.1250
Ecological risks of EDCs	0.5350	0.6188	0.7177
Ecological risks of heavy metals in sludge	–	–	0.0800

where e_i is the quotient value of the *i*th index, k is the constant that is related to the total number of evaluation objects, n is the total number of evaluation objects, w_i is the quotient weight of the *i*th evaluation index, and the sum of the quotient weights of all evaluation indexes is 1. The weight vector obtained in this study can be expressed as $W = (w_1, w_2, \dots, w_m)$.

For the most commonly used weighted average operator $M(+, \cdot)$, the fuzzy comprehensive evaluation was performed using Eq. (15).

$$B = W \times R = [w_1, w_2, \dots, w_m] \times \begin{bmatrix} r_{11} & \dots & r_{1n} \\ \vdots & \ddots & \vdots \\ r_{m1} & \dots & r_{mn} \end{bmatrix} = (b_1, b_2, \dots, b_m) \quad (15)$$

where B is the calculated result, W is the weight vector, R is the single-factor fuzzy evaluation matrix.

3. Results and discussion

3.1. Index weight

The weightings of the environmental impact indexes for the secondary processes, tertiary processes, and the entire procedures used in the WWTPs, determined using the quotient weighting method, are shown in Table 2. Higher weightings were generally assigned to the EIA indexes for ecological risks posed by EDCs than for the other risks and the other pollutants. The weightings for GHG emissions and the eutrophication potentials were different for different processes.

3.2. EIA results

3.2.1. GHG emissions

As shown in Fig. 2(a), GHG emissions during secondary treatment processes were primarily CO₂ emitted because of energy consumption and CO₂, CH₄, and N₂O emitted during the biological treatments. Differences in GHG emissions from the different WWTPs were mainly caused by different amounts of CO₂ being produced during the biological treatment processes. Particularly, BOD oxidation and endogenous respiration contributed large proportions of the total amount of CO₂ produced, and together contributed >81% of GHG emissions during the biological treatments. N₂O production contributed similar proportions (~10%) of total GHG emissions during the biological processes in all five WWTPs. CH₄ production contributed 5%–8% of total GHG emissions during the biological processes, i.e., less than the contributions of CO₂ and N₂O production.

Similar results were found by Yan et al. (2014) in a field study of gaseous and dissolved CO₂, N₂O, and CH₄ production in primary sedimentation basins, biological reaction basins, and secondary sedimentation basins in three WWTPs. In that study, CH₄ contributed 3%–7% of total GHG emissions from the biological reaction basins, and the contributions of CO₂ and N₂O were higher. However, slightly more N₂O was released in the study performed by Yan et al. than in this study, and N₂O production contributed 17%–38% of total GHG emissions (Yan et al., 2014). This may have been because a N₂O conversion factor was used in the Bridle model in this study, and the default conversion factor was lower than the actual monitored value. Nguyen et al. (2019) found that the aerated zone was the main source of GHG emissions

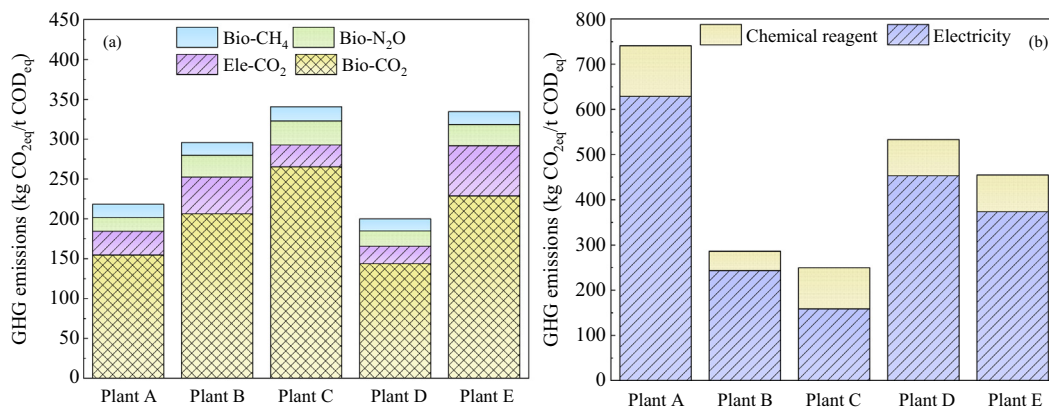


Fig. 2. The calculated GHG emissions of the (a) secondary and (b) tertiary processes.

during A²O processes and sequencing batch reactor treatment processes. They demonstrated that decreasing GHG emissions depended on controlling the dissolved oxygen concentration and aeration rate.

With respect to different WWTPs, the secondary treatment processes in Plants A and D produced smaller amounts of GHGs, with 200 and 218 kg of CO₂/t COD_{eq}, respectively, as a result of the high pollutant removal efficiencies of the processes in Plants A and D. In comparison, the secondary treatment processes in Plants B and C produced large amounts of GHGs because they had relatively low pollutant removal efficiencies. In addition, the UCT process in Plant E also produced large amounts of GHGs up to 335 kg CO₂/t COD_{eq}, mainly because of the amount of electricity consumed (which contributed 19% of the total amount of GHGs produced).

As shown in Fig. 2(b), GHG emissions during the tertiary treatment processes were mainly caused by electricity consumption, and chemical consumption was the next most important cause. The amounts of GHGs emitted were different for the different WWTPs because of the different tertiary treatment techniques used.

For example, the D-type filters in Plant A used large amounts of electricity and chemicals but had relatively low pollutant removal efficiencies, so larger amounts of GHGs were emitted by Plant A (740.6 kg CO₂eq/t COD_{eq}) than by the other plants.

3.2.2. Eutrophication potentials

As shown in Fig. 3(a), the eutrophication potentials for the secondary treatment processes were mainly caused by TP and NO₃-N emissions, which contributed 31%–52% and 28%–37%, respectively, of the total eutrophication potentials. The COD and NH₄-N concentrations in

the secondary treatment effluents made small contributions to eutrophication, 9%–18% and 3.4%–18.6%, respectively, of the total eutrophication potentials. Gallego et al. (2008) performed a study of the eutrophication potentials of emissions from 13 WWTPs using a functional unit of the production of 1 m³ of treated effluent. They found that the PO₄-P and NH₄-N concentrations in the effluent made large contributions to the eutrophication potentials of the water bodies receiving the effluent. Garrido-Baserba et al. (2014) determined the eutrophication potentials of effluents from 22 full-scale WWTPs in Spain and found that eutrophication was primarily caused by TN and TP in the effluents. Nitrogen and phosphorus concentrations in secondary treatment effluent are therefore of great concern in terms of potential eutrophication.

The secondary treatment processes in Plant C gave a higher eutrophication potential, of 3.3 kg PO₄-P_{eq}/t COD_{eq} compared with the secondary treatment processes in the other plants. It has been suggested that improving the nitrogen and phosphorus removal efficiencies will decrease the eutrophication potentials of the water bodies receiving WWTP effluent. It can be seen from Fig. 3(b) that the NO₃-N concentrations in the treated effluents were mainly responsible for the eutrophication potentials given by the tertiary treatment processes, contributing 78% of the total eutrophication potentials. TP in the effluent contributed 6%–11% of the total eutrophication potentials. The tertiary treatment processes coagulation/flocculation and filtration removed little TN but could remove 56%–77% of the TP. As shown in Figs. 3(b) and S1, smaller amounts of the pollutants, particularly TN and TP, were removed during the tertiary treatments in Plant A than during the tertiary treatments in the other WWTPs. Hence, Plant A therefore gave the highest total

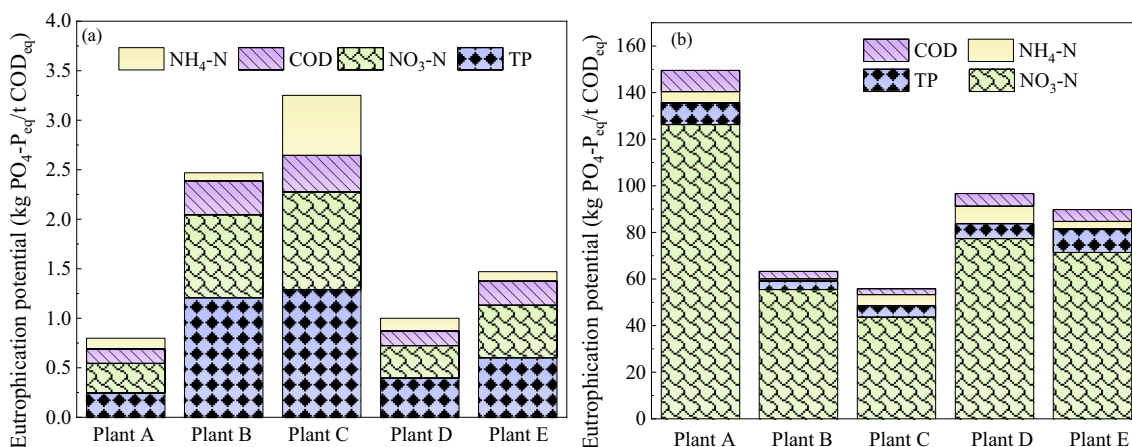


Fig. 3. The eutrophication potentials of the (a) secondary and (b) tertiary processes.

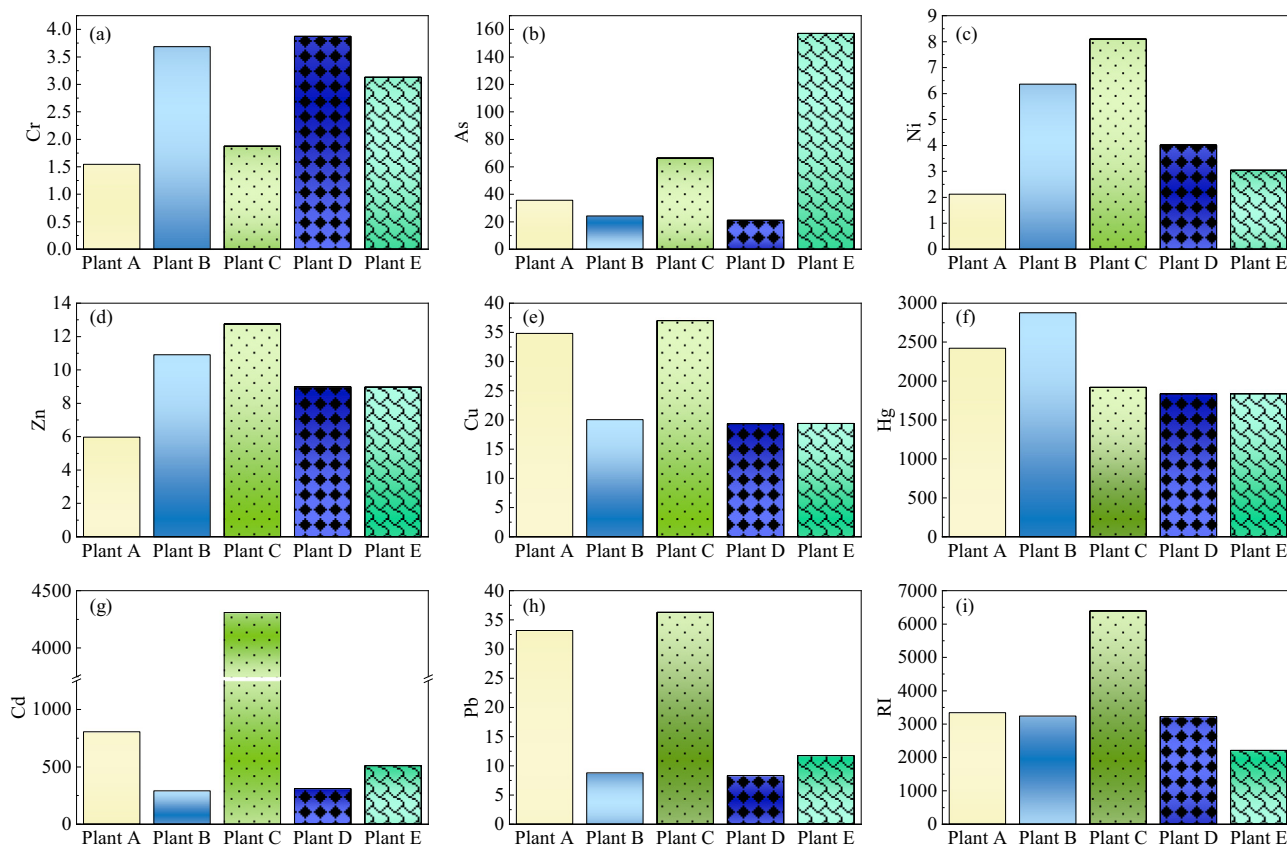


Fig. 4. The ecological risks of heavy metals in sludges of the WWTPs.

eutrophication potential, 150 kg PO₄-P_{eq}/t COD_{eq}. Larger amounts of pollutants, particularly COD, TN, and TP, were removed during the tertiary treatments in Plant C than during the tertiary treatments in the other WWTPs. Plant C gave the lowest eutrophication potential, 56 kg PO₄-P_{eq}/t COD_{eq}. Plant B also gave relatively high TP and COD removal efficiencies and an overall eutrophication potential of 63 kg PO₄-P_{eq}/t COD_{eq}.

3.2.3. Ecological risks of heavy metals in sludge

The potential ecological risks posed by the heavy metals in sludge produced by the WWTPs are shown in Fig. 4. The potential ecological risk indexes for Cr, Cu Ni, Pb, and Zn in the sludge produced by all of the plants were <40, indicating that the potential ecological risks

were low (Table S5). The ecological risks posed by As were very different for the sludge produced by the different plants, being low for As in sludge produced by Plants A, B, and D, moderate for As in sludge produced by Plant C, and considerable for As in sludge produced by Plant E. The Cd and Hg concentrations were markedly higher than the concentrations of the other heavy metals in the sludge produced by all of the plants, and the ecological risks were high for Cd and Hg in sludge produced by all of the plants. The ecological risk index for Cd was much higher for sludge produced by Plant C than for sludge produced by the other plants. The sums of the ecological risk indexes for the eight heavy metals were >2000 for all five WWTPs. Heavy metals in sludge probably pose high ecological risks, so sludge disposal needs to be managed appropriately. In comparison, Yang et al. (2017) evaluated

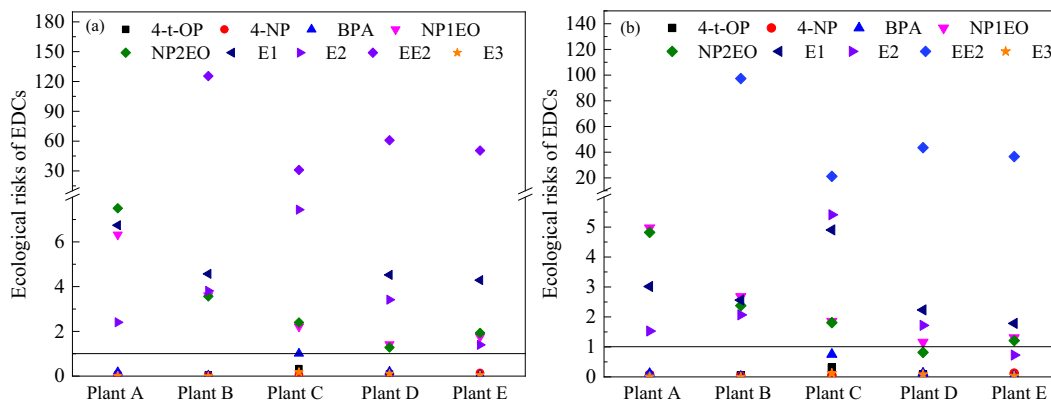


Fig. 5. The ecological risks of EDCs of the (a) secondary and (b) tertiary processes.

the potential ecological risks posed by Cd, Cr, Cu, Ni, Pb, and Zn in sludge produced by four WWTPs in Nanchang, China. They found total ecological risk indexes for the heavy metals of 4200–7500, suggesting that heavy metals posed high ecological risks. In particular, Cd posed a very high risk and was found to be likely to cause serious adverse effects. However, the background heavy metal concentrations in sludge, sediment, and soil can vary greatly because of differences in lithogenic effects in different areas.

3.2.4. Ecological risks of EDCs

As shown in Fig. 5(a), the ecological risk quotients for 4-t-OP, 4-NP, BPA, and E3 in the secondary treatment effluents produced by the five WWTPs were all <1, meaning these EDCs posed low ecological risks. However, the ecological RQs for NP1EO, NP2EO, E1, and E2 were >1, indicating that these EDCs probably posed high ecological risks and needed to be properly managed during secondary treatment processes. The total RQ was higher for the secondary treatment effluent from Plant B (RQ 141) than for the secondary treatment effluents from the other plants, and the RQs for the secondary treatment effluents from Plants A and C were relatively low (RQs 23 and 53, respectively). EDCs in secondary treatment effluents from Plants A and C were therefore considered to pose lower ecological risks than EDCs in secondary treatment effluents from the other plants. Controlling sources and secondary treatments are key strategies for controlling and decreasing the concentrations of EDCs such as NP1EO, NP2EO, E1, and E2 in effluent. Huang et al. (2014) compared the EDC removal efficiencies for four secondary treatment processes, an A²O-membrane bioreactor (MBR), an A²O system, an intermittent cycle extended aeration process, and an oxidation ditch. Better EDC removal performances were found for the A²O-MBR and A²O system than for the ICEAS and oxidation ditch.

The ecological risk quotients for EDCs in tertiary treatment effluents from the five WWTPs are shown in Fig. 5(b). The RQs for 4-t-OP, 4-NP, BPA, and E3 were <1 for all of the plants, indicating low ecological risks. However, the RQs for NP1EO and E1 in the effluents from all of the plants were >1, indicating that attention should be paid to removing these EDCs during tertiary treatments. The NP2EO, E2, and ethinyl estradiol RQs were > 1 for some of the WWTPs. The highest total RQ for the tertiary treatment effluents was for Plant B (RQ of 107), and relatively low total RQs were found for Plants A and C (RQs of 15 and 36, respectively). Tertiary treatment processes remove EDCs to some extent, but secondary treatment processes are the primary contributors to EDC removal from wastewater (Huang et al., 2014). Secondary treatment processes could be important for decreasing the ecological risks posed by EDCs in tertiary treatment effluent.

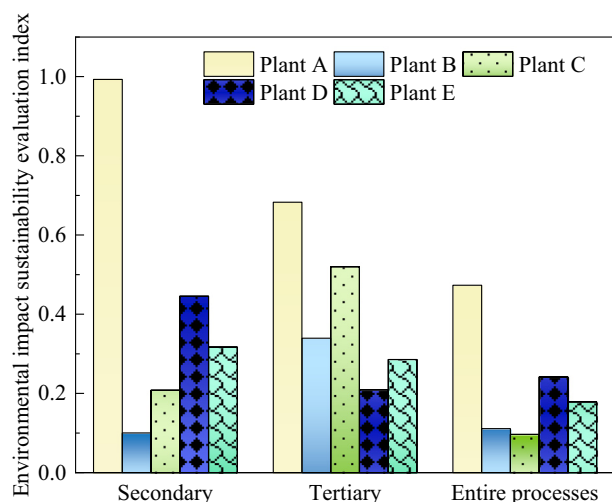


Fig. 6. The environmental impact sustainability evaluation indexes of different processes.

3.2.5. Fuzzy comprehensive evaluation

The secondary treatment processes in Plant A gave a comprehensive EIA sustainability index of 0.99, which was much higher than the comprehensive EIA sustainability indexes for the other WWTPs (Fig. 6). The weighting factors assigned to the different environmental impact indexes could have partly caused the differences in the sustainability indexes. The weightings given to the ecological risks posed by EDCs and the eutrophication potentials, 0.54 and 0.39, respectively, were higher than the weightings for the other parameters. The secondary treatment processes in Plant A gave good EDC and nutrient removal efficiencies, so Plant A had a high score in the comprehensive EIA sustainability evaluation. The sustainability index was higher for the Plant A tertiary treatment processes, 0.68, than for the tertiary treatment processes in the other plants because of the high EDC removal efficiency of the Plant A tertiary treatment processes. The sustainability index was also high, 0.52, for the Plant C tertiary treatment because of low GHG emissions and eutrophication potentials.

The comprehensive EIA sustainability indices for the entire WWTP procedures were mainly affected by the weightings of the different environmental impact indices and the technical performances of the plants. The quotient weightings for GHG emissions, the eutrophication potentials, and the ecological risks posed by EDCs were 0.0750, 0.1253, and 0.7195, respectively. Overall, Plant A performed well in terms of EDC removal and produced secondary and tertiary treatment effluents that posed low ecological risks, whereas Plant D gave the lowest GHG emissions and eutrophication potentials for the secondary treatment effluent. As shown in Fig. 6, the highest comprehensive EIA score was for Plant A (0.47) and the second highest was for Plant D (0.24).

4. Conclusions

A comprehensive method for evaluating the environmental impacts of treated effluents produced in WWTPs was developed. Biological treatments and electricity consumption were found to be the main contributors to GHG emissions, together contributing >60% of total GHG emissions. The eutrophication potentials indicated that high NO₃-N concentrations in effluent need to be decreased during the treatment processes. The ecological risk potentials indicated that special attention should be paid to removing EDCs (e.g., NP1EO and E1) and heavy metals (e.g., Hg) during secondary treatments and sludge treatments, respectively. The hybrid fuzzy evaluation allowed different WWTPs to be compared from various environmental perspectives objectively and rationally. The fuzzy results indicated which pollutants need to be removed more effectively than the current situation during the treatment processes. Differences in fuzzy membership functions and quotient weightings for different cases will indicate different priorities. Overall, the comprehensive evaluation approach can be used to identify and optimize key processes to mitigate the environmental impacts of WWTPs. The results will be useful for other current and future projects aimed at improving long-term environmental sustainability for WWTPs.

CRediT authorship contribution statement

Zhuo Chen: Data curation, Formal analysis, Writing – original draft, Funding acquisition. **Dan Wang:** Conceptualization, Methodology, Writing – review & editing. **Guohua Dao:** Resources, Investigation. **Qi Shi:** Resources, Investigation. **Tong Yu:** Resources, Investigation. **Fang Guo:** Resources, Investigation. **Guangxue Wu:** Conceptualization, Supervision, Project administration, Writing – review & editing.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.148212>.

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