REVIEW OF TOXICITY EMISSION FROM MUNICIPAL WASTEWATER TREATMENT BY LIFE CYCLE ASSESSMENT

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Abstract

Sewage treatment removes contaminants in wastewater, however pollutants from wastewater could also be transmitted to air, water and soil which could lead to negative impacts on the environment. The overall environmental impact from wastewater treatment plants (WWTPs) is very challenging to evaluate because conventional assessment tool such as environmental impact assessment (EIA) did not consider overall processes or input and output of the treatment plant. Therefore, a holistic method such as life cycle assessment (LCA) is needed to analyze the impact of WWTP's operation on the environment. This paper reviews the toxicity impact of WWTPs by LCA. The importance of toxic micropollutants impact from WWTPs will be discussed. Furthermore, studies regarding different life cycle impact assessment (LCIA) methodology used for toxicity impact assessment will be reviewed. Finally, a comparison on toxicity impact studies by LCA in developing countries also will be review. This review found that there is lack of studies concerning life cycle assessment that includes both metals and PPCPs contents in WWTPs especially in developing countries. Thus it is important to investigate higher number of micropollutants and other life cycle toxicity impacts from wastewater treatment plants using different LCIA methods especially in developing countries' situations to provide valuable info for LCA practice.

Keyword: Life Cycle Assessment, Wastewater treatment plants, Toxicity assessment, Pharmaceuticals and personal care products, Heavy metals

Introduction

The toxicity effect from sewage treatment plants has received high attention nowadays especially when new emerging pollutants are detected from municipal wastewater. Although toxic pollutants such as metals are detected in low concentrations, their increasing discharge from wastewater effluent could affect long-term threats to the environment (Bolong et al., 2009; Alfonsín et al., 2014). Thus, evaluating the toxicity effect from sewage treatment plants is significant to determine the risks from micropollutants and other priority pollutants.

At present, the impact of a wastewater treatment system can be evaluated by different evaluation tools such as the life cycle assessment (LCA) method, economic and exergy analysis (Muga & Mihelcic, 2008), the environmental impact assessment (EIA) method, and net environmental benefit analysis (NEBA). Particularly, LCA is an approach or method in assessing the environmental impacts associated with all stages in the life cycle of commercial products, processes or services. In LCA, environmental impacts are measured from raw material extraction of the product/process to the final removal of the materials, i.e cradle to grave (Li et al., 2013). Several software have been developed including free and commercial software to assist in the analysis of LCA. At present, various types of commercial LCA software are available such as SimaPro (El-Sayed et al., 2010), Gabi7 (Tomei et al., 2016), and Umberto. The structured methodology in LCA as stated in ISO starts with describing the goal and scope followed by life cycle inventory (LCI), life cycle impact assessment (LCIA) and finishes with a results interpretation is shown in **Table 1**. This methodology highlights the general steps of LCA with general characteristics that have been identified within each step.

Table 1 LCA methodology steps for environmental impact assessment from WWTPs

Goal and scope	Life cycle inventory	Life cycle impact assessment	Interpretation
•Objective •System boundary •Functional unit (e.g., 1m^3 of wastewater)	•Input data (e.g., influent, energy and chemical consumption) •Output data (e.g., emission to air, water and soil)	•Classification (e.g., eutrophication, global warming, acidification, ozone depletion, human toxicity, freshwater ecotoxicity and resource depletion potentials in midpoint impacts •Methodology selection (e.g., CML-IA, EDIP, IMPACT 2002+, eco-indicator99, Recipe and USEtox)	•Comparison of impact analysis •LCA method evaluation •Data quality/sensitivity analysis •Normalisation and weighting (optional)

(Source: JRC European commission, 2011)

This paper aims to review and evaluate the present state of knowledge with regards to environmental effects of toxic pollutants from wastewater treatment by LCA. This review will summarize and analyze published literature focusing on micropollutants emission and comparison of toxicity impact of different methods in LCA.

The importance of micropollutants

Rapid development and human activities lead to a rise of harmful elements in wastewater, making the urbanized areas a key passageway for metals and other toxic pollutants to the environment. Referring to the European Economic Community 1991 (EEC, 1991), the sewage treatment process contributes a significant amount of direct pollutants to the environment from sludge and effluent that contain toxic substances such as metals and micropollutants. The emission of metals from WWTPs consists of direct and indirect pollutants from electricity consumption, chemical consumption, effluent, and sludge. The direct metals in sewage such as mercury, copper, nickel, lead, and zinc mostly come from industrial and domestic wastewater, as well as rainwater runoff that enters the sewer system and leads to WWTPs (Ustun, 2009). These metals will eventually reach the environment from the effluent and sludge. The other indirect source of heavy metals are from electricity production such as barium, hydrogen fluoride, and nickel, which cause toxicity in humans by air or water contamination.

In addition, pharmaceuticals and personal care products (PPCPs) are micropollutants that also enter the environment after passing through sewer lines and WWTPs. Classes of pharmaceuticals include hormones, antibiotics, beta-blockers, and antidepressants. While, four classes of personal care products are found: fragrances, preservatives, disinfectants, and sunscreen agents. Most of the PPCPs, such as triclosan, 17a-ethinylestadiol, 17b-estradiol, and bisphenol-A, have been found at different levels of concentrations. In recent years, a few studies (Rosal et al., 2010; Kim & Farnazo, 2017; Rashid & Liu, 2020) have been conducted to determine the behaviour of these micropollutants in domestic and industrial wastewater, including surfactants, personal care products, pharmaceuticals and endocrine disruptors from the wastewater treatment process. Among these pollutants, pharmaceutical compounds have been identified as a great concern to surrounding communities as no legal standards have been set for their discharge into surface waters (Kim & Farnazo, 2017). For instance, recent investigations found that the concentration of pharmaceutical compounds in raw wastewaters (i.e., antibiotics, anti-inflammatories, hormones, and analgesics) vary greatly, resulting in inconsistencies in their behaviour during the treatment steps and their removal efficiencies.

These micropollutants are bioactive contaminants that cannot be fully eliminated with traditional wastewater treatment and are released daily in wastewater. Bolong et al. (2009) pointed out that these toxic substances are released back to the environment from effluents or adsorbed to the sludge at an average of 65%, depending on their lipophilic characteristics (i.e., the ability of compounds to dissolve). In 2000, the EU framework directive found 33 significant micropollutants in the aquatic environment comprising cadmium, lead, mercury, and nickel.

Meanwhile, emerging pollutants such as PPCPs have been described as the generation of new pollutants into the environment in significant amounts with harmful effects on organisms due to their abundant nature, persistence, bioactive and toxic characteristics in the environment. For instance, potential pharmaceuticals such as carbamazepine, diclofenac, and ibuprofen are considered as priority PPCPs for environmental monitoring because of their persistence formation in the water body, and possible contribution towards adverse human health effects (Archer et al., 2017). It is because most of the PPCPs are not biodegradable and cannot be treated by conventional wastewater treatment. Furthermore, the continuous discharge of micropollutants from wastewater effluent can cause long-term threats because the contaminants could form new toxic mixtures in the water body (Bolong et al., 2009; Alfonsín et al., 2014). Unfortunately, most of the current WWTPs, especially in developing countries, are not specifically designed to eliminate micropollutants (Gallego-Schmid et al., 2019). This is because monitoring action to these micropollutants or PPCPs has not been applied in most WWTPs due to discharge guidelines and standards that do not yet exist for most micropollutants.

Life cycle impact methods used for toxicity study

In LCA, the midpoint toxicity impacts have been classified into freshwater ecotoxicity, human toxicity, terrestrial ecotoxicity and marine ecotoxicity. The vital issue in toxicity impact categories is the uncertainty in the selection of the life cycle impact assessment (LCIA) methods (e.g., USES-LCA, EDIP, CML-IA, IMPACT2002 and USEtox) and its calculation tool. The various methodology varies significantly in terms of scope and modelling principles and can fail to attain the consistent characterization factors between each method. Thus, the choice of the most suitable LCIA method to toxicity impact is still uncertain (Renou et al., 2007).

CML-IA is the most commonly used methodology for LCA analysis of WWTPs, followed by EDIP2003. The CML-IA method considers a multi-media exposure, fate and effects model (Huijbregts et al., 2000). In CML-IA, human toxicity is considered, and ecotoxicity is separated into three impact categories: FEP, freshwater aquatic ecotoxicity; MEP, marine aquatic ecotoxicology; and TEP, terrestrial ecotoxicity. By contrast, the characterization of toxic effects in the EDIP2003 model is based on the independent fate, exposure, and effects model. EDIP2003 allows the LCA practitioner to compute toxicity potentials for human toxicity and ecotoxicity potentials, where human toxicity is allocated into three different exposure routes: HTP via air, HTP via water, and HTP via soil. Ecotoxicity is separated into three impact categories: acute FEP, chronic FEP, and chronic TEP. IMPACT2002+ method provides human toxicity (carcinogens and non-carcinogen), freshwater ecotoxicity and terrestrial ecotoxicity.

To harmonise the modelling methods that include characterisation factors of various life cycle toxicity impact, a life cycle initiative was announced by the United Nations Environment Program and the Society of Environmental Toxicology and Chemistry (UNEP-SETAC) in 2002 (Rosenbaum et al., 2008). In this programme, huge works were made to classify the causes of variances in toxicity-related models (Hauschild et al., 2013). Based on a range of existing LCIA methods (e.g., Impact 2002 and CML-IA), USEtox was established where infinite time is used as a sole time horizon (Rosenbaum et al., 2008). The USEtox model was provided based on the toxicity evaluation of pollutants and encompasses six emission sections: rural air, urban air, seawater, freshwater, agricultural land, natural land. It measures freshwater (aquatic) ecotoxicity and human toxicity with both cancer and non-cancer effects (Rosenbaum et al., 2008). As an outcome, USEtox was established and selected as a scientific consensus model after the evaluation among some models such as CalTox, IMPACT 2002, USESLCA, BETR, EDIP, WATSON, and EcoSenee for evaluating toxicity-related impacts in LCA (Rosenbaum et al., 2008; European commission, 2013). Nevertheless, due to the difficulty of determining characterisation factors, the CFs in USEtox are only interim instead of recommended for metals (Rosenbaum et al., 2008). Furthermore, available CFs for PPCPs in the existing USEtox model is very limited, and the modelling on fate, exposure, and impact pathways of chemicals is imprecise (Emara et al., 2018). By comparison, IMPACT 2002+ and USEtox are based on similar models, which demonstrate chemical fate, effect, exposure, and optionally severity model. The CML 2002/CML-IA is only differed by the calculation of effect and severity indicators. While, EDIP is a simplified method that estimates some of these processes without fully defining them (JRC European Commission, 2011).

Based on the available LCIA methods, some researchers use more than one method for the assessment in their project. Munoz et al. (2008) measured the possible environmental impacts on 98 priority and emerging pollutants using EDIP97 and USES-LCA methodology in WWTPs in Spain. They found that nickel is the priority pollutant in marine ecotoxicity potential using USES-LCA, whereas EDIP did not include this impact category. For further explanation on the LCA methodology for toxicity, Renou et al. (2008) assessed the influence of method selection by a case study of a full-scale WWTP in France. They concluded that there is a great difference of results from various LCA methodologies used that associated with toxicity impact categories. In this situation, not only the inventory of toxic substances but also the assessment methodology needs to be improved in LCA. Thus, the toxicity assessment of WWTP was suggested to identify whether the selection of the LCIA methods could affect the final result, strengthen the studied system and avoid a confusing decision (Li et al., 2019). This review concluded that the comprehensive methodology evaluation about toxicity impact from WWTPs containing both toxicity substances such as heavy metals and PPCPs is still lacking where the variabilities of toxicity substances in the wastewater, CF availability, and methodology choice could be the main impact on the final result.

Toxicity impact studies by LCA in developed and developing countries
Due to the importance of the toxicity effect from WWTPs, a few studies have started to evaluate the effect of toxic substances such as heavy metals and PPCPs on the environment, especially to human toxicity and freshwater ecotoxicity. Most studies related to WWTPs and toxicity impact originate from developed nations and to a lesser extent from developing countries (Lorenzo-Toja et al., 2016; Shimako et al., 2017; Emara et al., 2018). However, conclusions from these studies are not constant and comprehensive, and the summary of the studies is presented in **Appendix 1**.

In detail, Renou et al. (2007) found that one major issue in LCA toxicity impact research concerned large differences between the result from different life cycle impact assessment methods, mainly for human toxicity, but no detailed comparison on specific substances was made between the methods. Wenzel et al. (2008) conducted an LCA study of several wastewater treatment options. They considered the potential toxicity from heavy metals, endocrine disruptors, PAHs, phthalates, and detergents but only nine substances in total. Munoz et al. (2008) highlighted that PPCPs were relevant when evaluating the influent and effluent of WWTPs, but only PPCPs and direct heavy metals were measured in the study without doing a comparison with the indirect effect such as energy consumption. Li et al. (2019) found ecotoxicity impact results using the USEtox model increased by 25% after involving 126 PPCPs in life cycle inventory (i.e. based on secondary data in literature) of advanced wastewater treatment. Lorenzo-Toja et al. (2016) conducted a life cycle assessment considering records of heavy metals and PPCPs in WWTPs in Spain. The results showed no significant impact was found in the effluent when PPCPs were considered in the life cycle toxicity assessment using the CML 2002 methodology. However, they identified a significant effect of PPCPs (at an average increase of 40%) in the influent life cycle assessment scenario, highlighting that the impact of these micropollutants in untreated wastewater cannot be neglected. In addition, they mentioned the less of a scientifically vigorous scheme on which PPCPs emissions can be modelled, specifically during the end-of-life stage with limited coverage of active pharmaceutical ingredients in LCIA models.

Previous research shows that most of the toxicity impact categories were evaluated based on toxicity emissions from chemicals and electricity consumption in WWTPs such as the emission of sulphur dioxide and nitrogen oxides which contributes to human toxicity (Hospido et al. 2008; Piao & Kim., 2016). Besides this, impacts on terrestrial ecotoxicity are mainly due to the emission of heavy metals (e.g., zinc and copper) into the soil during the end-of-life of sludge. Kalbar et al. (2013) reported an almost similar result for 4 categories—human, freshwater, marine and ecotoxicity impact for four different types of WWT because they are not designed to remove heavy metals and other micropollutants. Thus, these previous studies considered normal operational parameters about the composition of the influent and effluent with only a few studies considering heavy metals and organic pollutants such as mercury and COD. In LCA, the presence of emerging pollutants in sewage are rarely considered due to the lack of local characterization factors representing environmental fate, exposure to humans and aquatic organisms, and toxic effects caused (Alfonsín et al., 2014). Thus, additional research is required to well characterize the consequences of micropollutants in the aquatic environment (Morera et al., 2016).

To analyze in detail the effect of these emerging pollutants, Lorenzo-Toja et al. (2016) conducted an assessment of heavy metals and PPCPs site measurement campaigns in Spain related to the winter and summer seasons along with the site-sampling of GHGs in two different units of WWTPs located in two different climatic regions, the Atlantic and the Mediterranean. The results for the toxicity impact-related categories indicated that similar performance was obtained in both regions, with winter being the most harmful season. However, a high concentration of heavy metals and PPCPs in the influents during summer

(57% higher than in winter) explains that there is a seasonal variation effect. Moreover, there has not been sufficient assessment of pharmaceutical pollutants in the environment from the Southeast Asian region with low strength wastewater. Thus, evaluation of production and usage of pharmaceutical products in all countries of Southeast Asia has been considered to be essential.

The volume of the pharmaceutical industry and human population in these countries has increased significantly in pharmaceutical contamination and its associated risk. For example, in Asia, the concentrations of antibiotics such as roxithromycin, trimethoprim, and sulfamethoxazole are high in both influent and effluent wastewater and surface water. Thus, the study of distribution and behaviour of PPCPs, as well as heavy metals, in the environment is crucial due to large quantities of its manufacturing however, little is known about this topic, especially in a tropical country such as Malaysia, Thailand or Indonesia. For example, as a developing country, Malaysia has seen a rapid development of better living conditions, leading to longer life expectancy and increased demand for pharmaceutical use at home or in the hospital. To date, some pharmaceuticals have been identified in the effluent samples from WWTPs in Malaysia, such as furosemide, metoprolol, salbutamol, mefenamic acid, atenolol and salicylic acid. Moreover, most of the previous toxicity studies for WWTPs were from developed countries with high strength sewage (e.g., COD value, 250-750 mg/L (Lorenzo-Toja et al., 2016). This highlights the lack of studies concerning both metals and PPCPs contents from WWTPs in developing countries with low strength wastewater (Rashid & Liu, 2020a), which could produce different environmental impacts. Furthermore, during wet weather times, domestic sewage that includes rainfall is the main component of urban wastewater influent to a WWTP. How the highly diluted water affects metals and PPCPs removal and the effluent concentration were barely discussed. This is especially true for tropical weather countries with high rainfall intensity, where sufficient data on this topic is not currently available.

Conclusion

From this review, it is shown that although most WWTPs met the local authority's regulatory requirements, many PPCPs and heavy metal compounds are still incompletely removed and later are discharged to the water stream and enter the environment in unknown amounts, especially in developing countries. This contrasts with the level of information about the effect of micropollutants from wastewater in LCA aspects already published and well documented in European and other developed countries with mostly high strength sewage. Therefore, further research is needed to investigate the occurrence of local organic pollutants, heavy metals, and PPCPs in WWTPs to identify their importance and contribution to provide valuable information for LCA practice. Overall, there is a need to improve this gap of knowledge in LCA specifically in the Southeast Asian region by investigating the impact of inclusion metals and PPCPs from WWTPs, as well as identifying the results from different LCIA methods.

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Conflict of interests

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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