Environment International 133 (2019) 105266

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Characterization of implementation limits and identification of optimization strategies for sustainable water resource recovery through life cycle impact analysis

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ARTICLE INFO

Handling Editor: Zhen Jason He

Keywords:
Resource recovery
Wastewater reclamation
Agricultural irrigation
Life cycle impact analysis
Optimization strategy
Environmental sustainability

ABSTRACT

How we manage alternative freshwater resources to close the gap between water supply and demand is pivotal to the future of the environment and human well-being. Increased scarcity of water for agricultural irrigation in semi-arid and arid regions has resulted in a growing interest in water reuse practices. However, insight into the life cycle impacts and potential trade-offs of these emerging practices are still limited by the paucity of systematic evaluations of different water reuse implementations. In this study, a host of environmental and human health impacts at three implementation levels of allowing water reclamation for crop irrigation was comparatively evaluated across the operational landscape via a combination of scenario modelling, life-cycle impact analyses and Monte Carlo simulations. Net harvesting of reclaimed water for irrigation was found to be dependent upon the sophistication of the treatment processes, since multistage and complex configurations can cause greater direct water consumption during processing. Further, the direct benefits of water resource recovery can be essentially offset by indirect adverse impacts, such as mineral depletion, global warming, ozone depletion, ecotoxicity, and human health risks, which are associated with increased usage of energy and chemicals for rigorous removal of contaminants, such as heavy metals and contaminants of emerging concern. Nonetheless, expanded simulations suggest the significance of concurrently implementing energy recovery, nutrient recycling, and/or nature-based, chemical-free water technologies to reduce the magnitude of negative impacts from engineered water reclamation processes.

1. Introduction

Over recent decades, water scarcity has been increasingly regarded as a global challenge because of population growth, economic development, and environmental degradation (Mekonnen and Hoekstra, 2016). By 2050, nearly 6 billion people in over 50 countries and regions will suffer from water poverty (Burek et al., 2016), and the quest to develop truly sustainable solutions to close the gap between water supply and demand has become one of the most pressing tasks that must be solved over the course of this century (Larsen et al., 2016). Reclaiming water from municipal wastewater provides a reliable solution for cities and regions with severe water scarcity. Much effort is being devoted to retrofit existing or develop new processes for water reclamation (Mihelcic et al., 2017; PUB, 2018), while the global budget for water reuse investment reached 12.2 billion US dollars in 2016 and may double by 2020 (BCCR, 2017).

Even with these opportunities, scientists, engineers and policy makers are increasingly concerned with the broader environmental and human health impacts of this emerging paradigm for enhancing water security. Recent viewpoints on the role of water reuse in achieving sustainability are contentious. Some researchers argue that irrigation water supply from reclaimed wastewater could mitigate the energy and

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https://doi.org/10.1016/j.envint.2019.105266
Received 23 July 2019; Received in revised form 2 October 2019; Accepted 13 October 2019
0160-4120/

carbon footprint associated with the exploitation of conventional freshwater resources, while reducing contaminant discharge into the environment and protecting downstream ecosystems (Cornejo et al., 2016; Wang et al., 2019). However, others claim a critical trade-off, namely the hypothesis that improving the quality of treated water might be achieved at the cost of high energy and chemical inputs alongside greenhouse gas (GHG) emissions across the life cycle, and in particular over the operational phase, of the system (Rahman et al., 2016; Remy et al., 2014). Still, others report that both the environmental benefits and burdens are vital clues to better develop, implement, and improve water resource recovery and end-use practices (Hao et al., 2019), and depend mainly on the system layout and local conditions (Bradford-Hartke et al., 2015; Hasik et al., 2017; Lu et al., 2018).

System design or treatment level implementation are significant factors that may play a key role in affecting the overall sustainability of water reuse services (Salgot, 2008). Globally, the most common action to implement water reuse is to directly divert the secondary effluent of wastewater treatment plants (WWTPs) for agricultural irrigation, partly because it requires few improvements to the constructed treatment trains and therefore alleviates the financial burden of upgrading and retrofitting existing infrastructure (Norton-Brandao et al., 2013). However, since most secondary WWTPs can also release numerous heavy metals (HMs) (Fu and Wang, 2011), contaminants of emerging concern (CECs) (Christou et al., 2017), and pathogens (Moazeni et al., 2017), sophisticated purification becomes necessary to safeguard public health prior to recycling of secondary effluents. For example, an increasing number of WWTPs have been upgraded by adding new treatment stages (such as chemical dosing) and/or integrated different secondary processes for the safe removal of more pollutants from wastewater for crop irrigation (Illueca-Munoz et al., 2008; Munoz et al., 2009). In addition, with the implementation of high-performance filtration technologies, such as reverse osmosis (RO) (McCarty, 2018; Tang et al., 2018), wastewater treatment systems can be customized in a more integrated fashion at the outset to enable delivery of high-quality reclaimed water for further reuse. Therefore, energy and chemical demands along with both direct and indirect environmental emissions can vary depending on the different levels of treatment implemented for water reclamation.

Recent studies have increasingly applied life cycle analysis (LCA) tools to explore, in part, the environmental impacts of wastewater reclamation (Baresel et al., 2015; Moretti et al., 2019; Opher et al., 2019; Pasqualino et al., 2011; Pintilie et al., 2016; Shiu et al., 2017; Tong et al., 2013). Nonetheless, most of these studies were focused on the environmental effects of specific system designs or one implementation approach. These studies neglected systematic evaluations of alternative water reuse implementations with identical contextual factors, and particularly without holistic investigation of potential solutions to reducing trade-offs while strengthening co-benefits through water reuse practices. To this end, insight into the life cycle impacts attributed to different wastewater reclamation implementations and optimization opportunities for sustainable water reuse practices are still limited.

In this study, a series of environmental and human health impacts attributed to three wastewater treatment implementation alternatives were comparatively assessed across the operational landscape using the LCA methodology. Alternate scenarios were built with the fixed purpose of using reclaimed wastewater for irrigating crops (Fig. 1), while Monte Carlo (MC)-based uncertainty analysis was included throughout the LCA to examine changes in impacts by varying the implementation levels of wastewater processing. Moreover, expansion of the primary system boundary was also pursued to investigate whether the negative impacts associated with water resource recovery practices can be reduced through proposed complementary solutions and to visualize the extent to which the potential magnitude of environmental credits can be benchmarked in comparison to the primary scenarios. Although the focus here is on three water reclamation schemes and the end-use of reclaimed water in agricultural activities, the approach and results of this study have broad implications of providing sustainable water services and integrated management of recovered resources, rather than merely delivering quantities of water.

2. Materials and methods

2.1. Overview of scenario modelling

Treated wastewater has been recognized as a viable freshwater resource for irrigation of agriculture in semi-arid and arid regions, as the single largest component of water consumption in such regions is for crop irrigation (Drechsel et al., 2015). In fact, the degree to which treated water can broaden the water supply portfolios of water agencies and substitute for other water supply sources depends heavily upon the composition of the effluent. Different water quality parameters could influence the suitability of reclaimed water for crops, including CECs, HMs, and pathogens, which can result in unwanted effects on human, plants, and soil health (Rahman et al., 2018). Nonetheless, specific water quality standards restricting these emerging contaminants in reclaimed water are still in their infancy. On the other side, treated water can provide appreciable amounts of essential nutrients (nitrogen and phosphorus) for plants via fertigation (Ren, 2019), yet the influence of the specific treatment train remains unclear. To evaluate and understand these diverse impacts, we proposed three primary scenarios to simulate the current water reuse practices representative in most regions of the world, where secondary and/or tertiary effluents are usually implemented for irrigating crop fields (Norton-Brandao et al., 2013; Zandaryaa, 2017). We made distinct assumptions in the individual scenarios regarding their municipal wastewater treatment levels and use of different treatment facilities capable of removing these contaminants, with the goal of exploring whether and how the level of implementation can influence the lifecycle impacts of water reclamation for agriculture.

As shown in Fig. 1, the diverting secondary effluents (DSE) scenario assumes that municipal wastewater is handled by secondary treatment facilities and then reused for crop irrigation. To ensure the representativeness of the current study, we included various common secondary treatment approaches, which consist of the Modified Ludzack-Ettinger, Bardenpho, oxidation ditch, sequencing batch reactor, and so on, were implemented in the DSE scenario. The retrofitting existing systems (RES) scenario assumes further increments in the removal efficiencies of waterborne contaminants in secondary effluents to allow safer irrigation of treated water for crops via equipping tertiary treatment processes or combining different secondary facilities. In brief, alternative tertiary configurations, such as chemical phosphorus removal and combinations of the above-mentioned secondary treatment approaches were included in the RES scenario. The customizing reclamation facilities (CRF) scenario tailors the implementation of municipal wastewater treatment to intended reuse at the outset, and is enabled by emerging high-performance treatment technologies, such as RO and other emerging methods. A summary of the treatment methods and process configurations selected for each scenario is provided in Supplementary Information (Table S1).

The above scenarios were all assumed to handle municipal wastewater with an influent flow of 1×10^4 m^3/d, chemical oxygen demand (COD) of 500 mg/L, total nitrogen (TN) of 50 mg/L, and total phosphorus (TP) of 12 mg/L. The main effluent quality parameters (COD, TN, and TP) were estimated based on the reported treatment efficiencies of each method under the scenarios (Table S1). Given the emerging significance and human health concerns in respect of CECs and HMs (Zandaryaa, 2017), the present analysis also includes parameters for the occurrence and removal efficiencies of a host of CECs and HMs throughout the treatment processes (Text Sections S1–S2 and Tables S2–S6). The existence of pathogens in effluents also becomes a key factor in influencing utilization of reclaimed water for crop

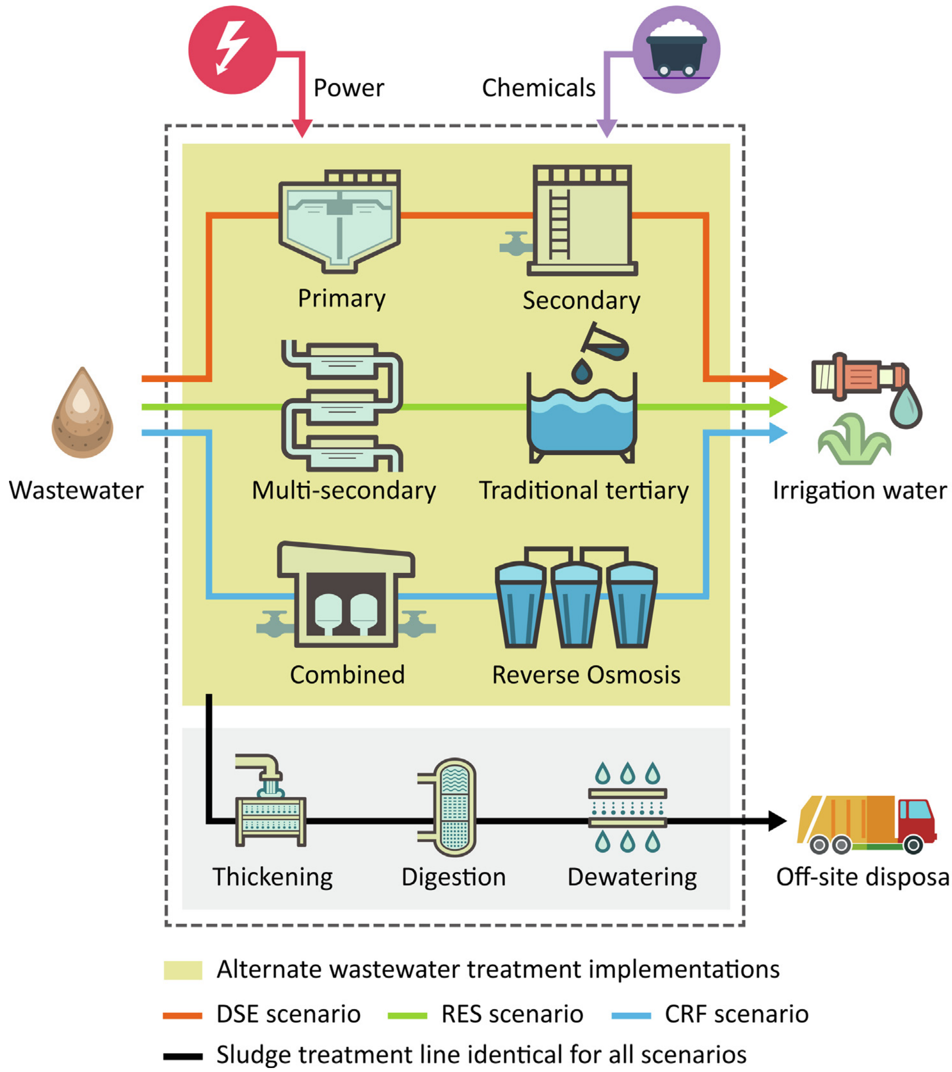

Fig. 1. Conceptual process flow diagram for building DSE, RES, and CRF scenarios. Implementing irrigation of reclaimed-water for crops avoids production of irrigation water and manufacturing of commercial fertilizers. Both are included within the system boundary of the analysis but are not shown in the diagram.

irrigation. Nevertheless, pathogens and their related risks are highly dependent on the specific local conditions of wastewater streams and country-level or even higher-resolution differences in water reuse regulations (EU, 2016). Moreover, common secondary and tertiary wastewater treatment approaches are documented as presenting comparable capacities in reducing pathogens (UN, 2017a; WHO, 2006). In addition, microbial hazards are commonly addressed in risk assessment studies, but are not routinely included in life cycle-based analyses (Harder et al., 2015). For these reasons, estimates of pathogens in effluents and their related impacts throughout the water reuse practices were excluded from this analysis.

2.2. *Life cycle goal and scope*

The aim of the present LCA is to comparatively analyze the environmental and human health impacts of varying levels of implementation of wastewater reclamation (DSE, RES, and CRF scenarios) and end-use of reclaimed water (for agricultural irrigation). The functional unit selected for the present study was 1 m^3 of wastewater treated and reused for irrigating crops, which allows the three primary scenarios to be compared systematically rather than solely in terms of wastewater treatment or end-use practices. A 20-year design life was considered because of the common lifespans of engineered water treatment infrastructure (Renou et al., 2008; Theregowda et al., 2016), and this was also employed in another study that presented similar reasons (Morera et al., 2017). The operational phases were included in the analysis, whereas the plant construction and decommissioning after the 20-year design life were excluded owing to their negligible effects in comparison to the long-term operations (Foley et al., 2010; Hao et al., 2019). Comparison of the DSE, RES, and CRF scenarios accounts for the life cycle impacts in two processes (Fig. 1): (1) foreground (wastewater treatment and reuse; excess sludge generation, anaerobic stabilization of sludge, and off-plant disposal; on-site gaseous waste generation and emission); and (2) background (energy, chemicals, and other materials required throughout the on-site and off-site foreground processes). It is noteworthy that the impacts associated with the transportation of reclaimed water from treatment plants to irrigation sites were excluded from the analysis, since the transport distances were assumed to be identical in all the primary scenarios. Further, the dynamics of nutrients (including fugitive emissions, crop intake, losses to the waters, and soil retention) throughout the irrigation of reclaimed water or land use of excess sludge were included to clarify the concurrent impacts of resource reuse for agricultural activities (Table S7). It should be noted that no specific crops were considered in this study. While considering the potential variations in uptake of nutrients by different plants, typical range values from literature were used to ensure the representativeness of the present analyses. The availability of effluent- and/or sludge-derived nutrients for crops was also assumed to replace commercial fertilizers (Bradford-Hartke et al., 2015), such that the mitigated environmental impacts relative to the production and utilization of fertilizers were also considered.

2.3. Life cycle inventory and data acquisition methods

A life-cycle inventory (LCI) or comprehensive list of inputs and emissions across the life cycle was compiled for each of the scenario designs. It is well known that the profiles of resource inputs and environmental emissions during wastewater treatment services are usually affected by the water quality parameters and process configurations applied (Mousel et al., 2017). Therefore, the total operating energy (LCI_e, kWh), chemical consumption (LCI_c, kg), and excess sludge production (LCI_s, kg) for a certain water treatment process configuration n (n = 1, 2, 3, … , N; N = 21 in this study, see Table S1) are calculated using the proposed methodology given below (Eq. (1)):

$$LCI_i^{(n)} = \sum_j (M_j^{(n)} \times w_{i,j}^{(n)} \times \varepsilon_{i,j}^{(n)}) \tag{1}$$

where subscript i distinguishes the different LCI categories (LCI_e, LCI_c, and LCI_s) and j represents influential waterborne pollutant (COD, TN, or TP). $M_j^{(n)}$ represents the removal of contaminant j in kg (Eq. (2)), $w_{i,j}^{(n)}$ stands for the unit value of LCI category i per kg of contaminant j removed (acquired from the literature and summarized in Tables S8–S9), and $\varepsilon_{i,j}^{(n)}$ represents an allocation parameter (dimensionless) for quantifying the significance of the contribution arising from the process of removing contaminant j (Eq. (3)) for each individual LCI category (LCI_e, LCI_c, and LCI_s).

$$M_j^{(n)} = \frac{Q \times C_j \times R_j^{(n)}}{1000} \tag{2}$$

where Q represents the total volume of wastewater treated per day (m^3; $Q = 1 \times 10^4$ m^3 in this analysis), C_j indicates the influent concentration of contaminant j (mg/L; in this study, C_j is 500 mg/L for COD, 50 mg/L for TN, and 12 mg/L for TP), whereas $R_j^{(n)}$ denotes the removal efficiency of contaminant j for process configuration n.

$$\varepsilon_{i,j}^{(n)} = \frac{\delta_{i,j}^{(n)}}{\sum_j \delta_{i,j}^{(n)}} \tag{3}$$

where $\delta_{i,j}^{(n)}$ represents the coefficient of variation of the value set of $w_{i,j}$ (Eq. (4)):

$$\delta_{i,j}^{(n)} = \frac{\sqrt{\frac{1}{N}\sum_{n=1}^{N}\left(w_{i,j}^{(n)} - \frac{1}{N}\sum_{n=1}^{N} w_{i,j}^{(n)}\right)^2}}{\frac{1}{N}\sum_{n=1}^{N} w_{i,j}^{(n)}}. \tag{4}$$

Direct emissions of organic matter (COD), nutrients (TN and TP), CECs, and HMs to aquatic, terrestrial and/or atmospheric systems via effluent and/or sludge disposal on land were estimated based on a combination of the following assumptions and parameters derived from existing studies (Fig. S1). Specifically, both biogenic and non-biogenic carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) on-site emissions to the atmosphere were estimated for wastewater and sludge treatment processes based on complementary procedures from the USEPA (2010) and a previous study (Law et al., 2013). The relevant determining factors are summarized in Table S10. Off-site fugitive emissions (CO_2, CH_4, N_2O, H_2S, and NH_3) from the irrigation of treated water (Table S7) and disposal alternatives of excess sludge were estimated based on reference emission factors (Table S11). It is noteworthy that although the irrigation approach was not customized in this analysis, a range of values were set to simulate potential variations in off-site fugitive emissions throughout different irrigation methods in real-life practices.

2.4. Life cycle impact assessment

The environmental and human health impacts of the scenarios were characterized using the Hierarchist ReCiPe 2016 mid-point based method, ver. 1.1 (Huijbregts et al., 2017). The impact categories of water use, resource scarcity, eutrophication, toxicity, acidification, global warming, ozone depletion, as well as human health risks (both carcinogenic and non-carcinogenic), were selected for the current evaluation. These parameters represent the local water quality, broad-scale impacts, and human health concerns most commonly included in integrated water management LCAs.

In particular, ecotoxicity and human health toxicity were estimated in the ReCiPe 2016 model using the Uniform System for the Evaluation of Substances adapted for LCA (USES-LCA), ver. 2.0 (van Zelm et al., 2009). The majority (90%) of the 181 CECs studied had existing characterization factors (CFs) already implemented for ecotoxicity and human health impacts in the USES-LCA model. The remaining CECs with unknown CFs were derived from previous experiments and substituted into the USES-LCA model for further assessment. Table S12 lists the CF values for the remaining 19 CECs that were evaluated in this manner, whereas the CFs of the other CECs were acquired from the basic data library of the USES-LCA model (not presented in this work). The water use category was estimated in the ReCiPe model based on water consumption embedded in background processes (manufacturing of energy and chemicals). The reclaimed water yield and direct water consumption during treatment processes were concurrently included in this study to closely fit the approach with the goal of the current analysis (referred to as the water resource recovery category). In addition, copper was substituted by phosphorus in the estimation of mineral resource scarcity potential using a conversion factor (0.167 kg P/kg Cu) (Huijbregts et al., 2017), since removal and recovery of phosphorus was more relevant to wastewater treatment services than that of copper (Bradford-Hartke et al., 2015). Subsequently, the modified ReCiPe model was accessed in the LCA platform SimaPro ver. 8.5 (PRé Sustainability), equipped with Ecoinvent database ver. 3.4, to determine the life cycle impacts. In addition, it is noteworthy that various electricity mix components were considered here for quantifying the impacts related to energy consumption throughout the processes. Thus, it was assumed that 70%, 19%, 3%, and 2% of the electrical energy were derived from hard coal, hydroelectric power, wind power, and a combination of nuclear and natural gas, respectively, building upon the Chinese electricity mix provided in Ecoinvent.

Notably, the LCI statistics of all process configurations in relation to each scenario were used as model inputs for subsequent calculation of the impacts in SimaPro. Therefore, the model outputs presented here are all related to the overall results of each individual scenario rather than the process configurations.

2.5. Uncertainty analyses

The statistical uncertainties in the LCI inputs were captured via MC analysis by random sampling (10,000 trials) from the underlying probability distributions obtained from public databases and/or previous work. All ranges and parameter values are given, where appropriate, in the Supplementary Information, presenting the main assumptions and variations in water consumption from treatment trains, embodied energy, chemical use, gaseous emission parameters, avoided production and application of fertilizers, and occurrences and removal of CECs and HMs.

3. Results and discussion

3.1. Water resource recovery potential

Our approach considers life-cycle water resource recovery potential for crop irrigation based on the balance of: (i) reclaimed water

Table 1
Life-cycle water use and net gain of reclaimed water for agricultural irrigation, expressed per cubic meter of wastewater treated.

Process	Life-cycle water use (median), m^3/m^3		Net gain of reclaimed water (median, 5th percentile, 95th percentile), m^3/m^3
	Direct[a]	Indirect[b]	
DSE scenario	0.04	1×10^{-3}	0.96, 0.96, 0.98
RES scenario	0.23	5×10^{-3}	0.76, 0.73, 0.82
CRF scenario	0.25	7×10^{-3}	0.75, 0.71, 0.79

[a] Includes direct water use and loss during wastewater treatment processes, which were median estimates of the relevant values in Table S13.
[b] Median water consumption from upstream power plants and chemical factories.

production; (ii) on-site water use and loss; and (iii) indirect water consumption from the generation and delivery of energy and chemical inputs for water and sludge treatment trains. As shown in Table 1, implementing water resource recovery in the three scenarios had identical potential to reuse nearly 100% of each 1 m^3 of wastewater entering the treatment facilities, meaning that indirect water consumed in upstream power plants and chemical factories can be negligible.

However, the median of direct water consumption was much higher for the sophisticated purifications of the RES or CRF scenarios than that encountered in implementing the more common wastewater treatments of the DSE scenario (0.23 m^3/m^3 or 0.25 m^3/m^3 vs. 0.04 m^3/m^3). The processes that require added freshwater use such as RO, or processes that involve multistage treatment with complex circulating stream pipelines, usually result in high-levels of water use and/or loss potential (Tong et al., 2013). In this study, median direct water consumption for sophisticated configurations that include multistage combinations of secondary processes or equipped with complex tertiary units was approximately six times greater than the single secondary processes of the DSE scenario (Table S13). These results indicate that the median life-cycle water resource recovery potential for crop irrigation can be increased by approximately 30% when the common DSE scenario is used instead of more sophisticated scenarios (RES and CRF). Nonetheless, much lower values of significant contaminants, such as CECs and HMs, were found in the reclaimed water of the RES and CRF scenarios than that of the DSE scenario (Table S14). Consequently, the quantity of water reclaimed for crops was negatively related with the water quality improvements provided by increasing the sophistication of treatment schemes. In other words, the direct advantages of water resource recovery realized by the use of advanced implementations can be reduced by the increase in direct water consumption associated with more rigorous purification processes.

3.2. Eutrophication and mineral resource scarcity potentials

Diverting effluent from the receiving water bodies to crop irrigation favors downstream ecosystems, considering its negligible effects on aquatic eutrophication (Wang et al., 2018). Despite this benefit, life cycle eutrophication potentials across water reclamation and reuse still present various indirect, positive, and/or negative eutrophication effects (Fig. 2a and b). The conventional option for water reuse (DSE scenario) had the highest freshwater eutrophication potential, of which the majority attributed to nutrient losses throughout effluent irrigation (Fig. 2a, grass green symbology). Phosphorus removal in the DSE scenario was achieved through traditional secondary processes, but much more influent phosphorus remained in the effluent (~60%; Fig. 2c) and was recycled via fertigation. This favored a reduction in life-cycle freshwater eutrophication potential, which agreed with previous analyses that greater indirect eutrophication potential accompanying the production and utilization of commercial fertilizers could be avoided through recycling of wastewater phosphorus (Bradford-Hartke et al., 2015). However, irrigation of phosphorus-rich effluent can concurrently cause increased eutrophication potential, as phosphorus loss to waters from fertigation was proportional to the amount of phosphorus in the effluent.

The sophisticated options (RES and CRF) were observed to reduce phosphorus loss to waters from fertigation (Fig. 2a and b). This could be attributed to the fact that chemicals (mainly iron chloride in this study) were added to rigorously remove phosphorus in the RES and CRF scenarios, in which the majority of the influent phosphorus accumulated in the excess sludge (> 90% for RES and ~100% for CRF; Fig. 2c). Meanwhile, chemical phosphorus precipitation was assumed to have effectively immobilized phosphorus within the excess sludge, thereby hampering phosphorus loss from disposal of iron-rich sludge, and thus reducing the unwanted impacts of eutrophication in the RES and CRF scenarios. Further, both electricity demand and chemical use for treatment processes to implement the CRF scenario were one-fold higher than those needed to operate the DSE and RES scenarios (Fig. 2a), demonstrating that complex configurations would also result in indirect burdens on freshwater eutrophication.

Moreover, the three wastewater reclamation scenarios were found to have slightly different median marine eutrophication potentials, although the scenarios could still be differentiated on the basis of accumulative potential (Fig. 2b). This was because the major proportion of influent nitrogen in all the primary scenarios had been biologically transformed into nitrogen gas (N_2) and emitted to the atmosphere (67%, 76%, and 91% for DSE, RES, and CRF, respectively; Fig. 2c), whereas the differences were dominated by changes in nitrogen loss to waters from the fertigation process. To date, the rates of nitrogen leaching from solids disposal on land are rarely reported differentiated into biological and chemical solids. Accordingly, the leaching rates of nitrogen were assumed identical for both biological and chemical sludge, and thus it is not surprising that the CRF scenario exhibited greater marine eutrophication potential attributed to disposal of nitrogen-rich sludge (Fig. 2b and c).

Direct use of conventional secondary effluents as an agricultural irrigation source also benefited from the net decrease in mineral resource depletion through the avoidance of mineral use in fertilizer production (Fig. 2d). In other words, phosphorus recovery along with crop irrigation by DSE effluent reached ~0.11 g P-eq minerals/m^3 of reclaimed water used. However, considering the relative depletion of phosphorus in the enhanced secondary and tertiary effluents from the RES and CRF scenarios (Fig. 2c), mineral requirements for electricity and chemical production substantially exceeded the avoided mineral use in the manufacturing of fertilizers, resulting in the median net depletion of 0.23 g P-eq/m^3 and 0.17 g P-eq/m^3, respectively (Fig. 2d).

3.3. Ecotoxicity and human health impacts

WWTP effluents are considered one of the major sources of CECs and HMs in receiving waters (Fu and Wang, 2011; Tran et al., 2018). Therefore, reclaimed water for agricultural uses might be associated with environmental risks and adverse health effects that must be assessed prior to implementation of large scale practices. The statistical ranges shown in Fig. 3a reflect the overall life cycle toxicity impacts across different scenarios and parameter uncertainties within each scenario. The Sankey-based analysis (Fig. 3b) isolated both human toxicity and ecotoxicity potentials for each scenario at the median level of 10,000 MC simulations expressed in kg of 1,4 DCB-eq/m^3 of wastewater reused. The highest median of total toxicity potential was captured in the DSE scenario (3.70 kg 1,4 DCB-eq vs 3.03 kg 1,4 DCB-eq in RES or 2.84 kg 1,4 DCB-eq in CRF), reflecting that direct irrigation of secondary effluent without rigorous removal of HMs and CECs, could lead to the highest adverse impacts on ecosystems and human health. It is noteworthy that the estimation of the overall toxicity was highly uncertain because the removal efficiencies of HMs and CECs varied widely across different treatment processes.

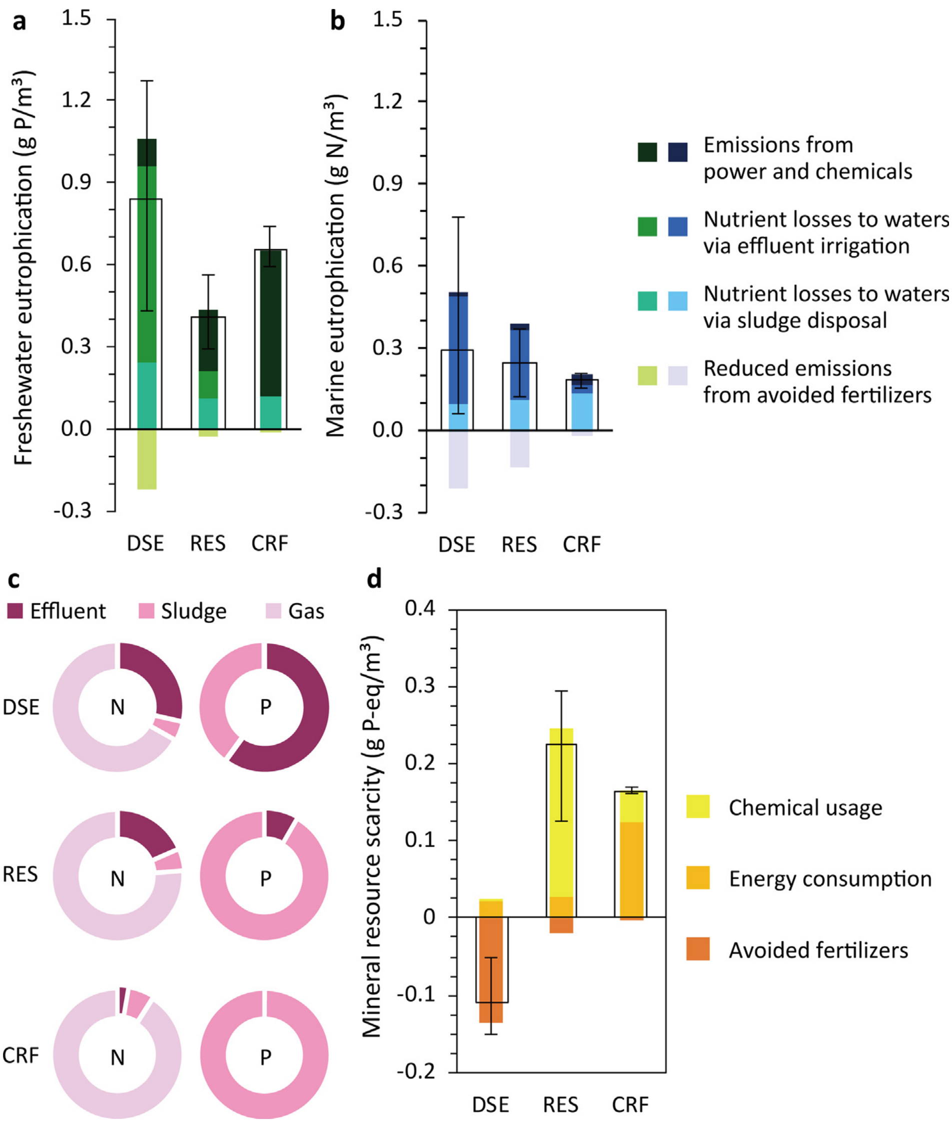

Fig. 2. Comparison of life-cycle freshwater (a) and marine (b) eutrophication potentials, accumulative balance of nutrients (c), and mineral resource scarcity (d) for the three investigated scenarios. The related size or the apparent absence of each colored bar segment represents either the median contribution of the process to each impact (for a, b, and d), or the proportional distribution of influent nutrients in different phases after treatment (for c) at the median level. The black frames in a, b and d arepresent the median net contributions of the entire scenario to each impact resulting from 10,000 MC simulations, whereas the error bars represent the respective 5th and 95th percentiles. A negative value indicates an environmental benefit, whereas a positive value indicates an increase in the environmental burden.

In the three scenarios explored, the diversion of treated effluent from the receiving water bodies to agricultural irrigation could significantly reduce the toxicity effects on both freshwater and marine ecosystems, producing aquatic toxicity impacts in the toxicity categories that were essentially negligible (Fig. 3b). However, the ecological toxicity effects on terrestrial ecosystems were significant and the approaches to treatment also influenced terrestrial ecotoxicity potentials. For instance, power plant derived emissions played a

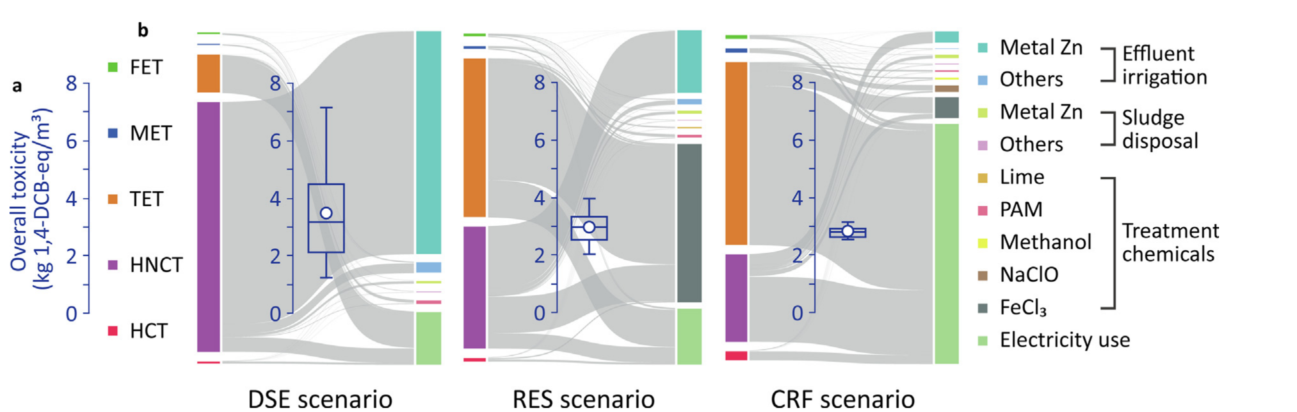

Fig. 3. Overall toxicity potentials for the three primary scenarios (a), and insight into ecological toxicity and human health, carcinogenic and non-carcinogenic toxicity contributions of each scenario's processes (b). The center lines in the box-plots represent median values, circles represent average values, boxes represent 25th and 75th percentiles, and bars represent 5–95th percentiles of the distributions resulting from 10,000 MC simulations. The median values of the simulations under each scenario were used to prepare the respective Sankey diagram. Abbreviations: FET, freshwater ecotoxicity; MET, marine ecotoxicity; TET, terrestrial ecotoxicity; HNCT, human non-carcinogenic toxicity; HCT, human carcinogenic toxicity.

predominant role in generating terrestrial ecotoxicity potential from the DSE scenario (nearly 0.28 kg 1,4 DCB-eq/m^3, 92% of the total), since no significant chemicals were needed to achieve advanced nutrient removal. In contrast, terrestrial ecotoxicity accounted for over half of the total toxicity for the RES scenario (~1.48 kg 1,4 DCB-eq/m^3), in which the manufacture of iron chloride for chemical phosphorus precipitation was the highest contributor to the impacts, whereas emissions from the production of electricity needed for water and sludge treatment lines within WWTPs accounted for the second largest contribution. This result is very consistent with the assumption that chemical precipitation is integrated with the secondary treatment processes in the RES scenario for improved phosphorus removal, which was also in agreement with the previous outcome that chemical-driven phosphorus removal could result in greater terrestrial ecotoxicity (Rahman et al., 2016). Treatment alternatives that utilize RO techniques (in the CRF scenario) had a comparable level of life cycle terrestrial ecotoxicity to treatment approaches in the RES scenario (1.72 kg 1,4 DCB-eq/m^3 and 1.48 kg 1,4 DCB-eq/m^3, respectively). However, they resulted in higher terrestrial ecological toxicity as the RO processes showed at least fourfold and fivefold increases in energy consumption compared to those configurations used in the DSE and RES scenarios, respectively (Table S13).

Given the existing and emerging differences in exposure pathways, modes of action, and types of effects, human health related toxicity is now usually assessed in separate carcinogenic and non-carcinogenic impact categories (Gifford et al., 2018). As shown in Fig. 3b, human health risks significantly decreased with treatment implementation levels, whereas the contributions of carcinogenic impacts to life cycle human toxicity were apparently minor (< 1%, < 3%, and < 10% for DSE, RES, and CRF scenarios, respectively, without considering uncertainties) compared to those of non-carcinogenic human health impacts. In the DSE scenario, ~90% of non-carcinogenic impacts were generated through agricultural usage of reclaimed water, in which effluent-derived zinc (Zn) with additional HMs and CECs were the predominant contributors to the impacts. The contributions of HMs and CECs to non-carcinogenic emissions were significantly diminished (particularly for the CRF scenario), because the removal efficiencies of waterborne contaminants were higher in the RES and CRF scenarios. However, the contribution hotspot shifted from wastewater reuse to indirect emissions associated with the upstream manufacturing of chemicals and energy required for the treatment processes. This indicates that although sophisticated processes, such as RO, indeed reduced contaminant concentrations in the reclaimed water and lessened the relevant non-carcinogenic impacts at the site of the application, their implementation may actually lead to increases in indirect release of non-carcinogenic emissions from upstream energy and chemical plants.

3.4. Global warming, ozone depletion, and acidification potentials

Different treatment implementation schemes for wastewater reclamation and reuse can also lead to variations in other broader environmental effects, including life cycle global warming, ozone depletion, and terrestrial acidification, resulting from direct WWTP emissions and indirect upstream emissions. Accordingly, Fig. 4a depicts the global warming potential of the three implementation scenarios. Direct emissions of CH_4 from onsite treatment lines within WWTPs and off-plant sludge disposals were found to be remarkable sources of GHG emissions in all the scenarios. However, additional fugitive emissions from the CRF scenario increased the median global warming impacts by over 48% and 86% compared to those of RES and DSE scenarios, respectively. This arises because processes that are clearly electricity intensive, such as RO (results presented in Table S13), are usually associated with greater levels of indirect GHG emissions from the generation of electrical energy, which is in agreement with previous work (Rahman et al., 2016). This indicates that although improved quality of reclaimed water was achieved at the cost of either treatment

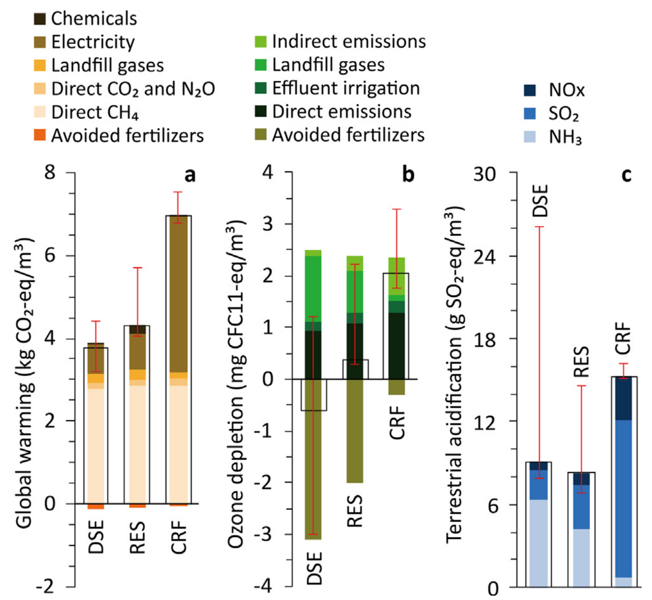

Fig. 4. Global warming, ozone depletion, and terrestrial acidification potentials for different scenarios. The error bars represent the 5–95th percentiles, whereas the transparent boxes represent average values from 10,000 MC simulation trials.

chemicals or embodied energy in the present analysis, the processes that need additional chemicals rather than electrical energy to improve water treatment can have significantly lower global warming potential than electricity-intensive processes, such as RO.

Moreover, approximately 90% (neglecting uncertainties) of the ozone depletion potential is attributed to onsite emissions from water and sludge treatment processes (Fig. 4b), particularly from the release of fugitive N_2O. It should be noted that N_2O production and emissions during the treatment processes were estimated based on the amount of nitrogen removed. To this end, it is not surprising that the implementation of rigorous nitrogen removal processes resulted in elevated ozone depletion effects from N_2O releases in the CRF scenario. Diverting the effluent to crop irrigation recycled nitrogen concurrently to plants, and eliminated the highest amount of fugitive N_2O emissions from the production of fertilizers in the DSE scenario, leading to net reduced ozone depletion potential generated in the DSE scenario. This is due to the relatively lower proportion of wastewater nitrogen decomposed into N_2 with conventional secondary processes, whereas the nitrogen retained in the effluent was therefore highest in the DSE scenario. It is noteworthy that the high uncertainties exhibited in the life cycle ozone depletion impacts were dominated by existing large variations in factors reported for estimating N_2O releases.

The CRF scenario had the highest terrestrial acidification potential over the range of variability and uncertainty explored (Fig. 4c), indicating that acidifying substances released from upstream fossil fuel combustion (SO_2 and NO_x) are the essential drivers of the acidification impacts. This can again be explained by the fact that energy intensive processes (e.g., RO) were implemented in the CRF scenario.

3.5. Optimizing the implementation methods to minimize unwanted effects

The significant impacts of energy and chemical inputs for engineered wastewater reclamation processes and environmental emissions from wastewater and sludge handling suggest that there may be opportunities to minimize the negative effects by considering the potential to implement energy recovery, reduced chemical usage, and/or agricultural application of the remaining nutrients. Both N_2O and CH_4 are critical GHGs that are approximately 265 and 28 times more powerful than CO_2, respectively (Lee et al., 2014). They are also both common sources of energy in numerous applications in which power is produced using N_2O as an oxidant in CH_4 combustion (Scherson and Criddle, 2014). Therefore, N_2O and CH_4 generated from bioprocesses were assumed to be captured and co-combusted as power for the treatment plants (referred to as the Energy Recovery Solution). In addition, nature-based solutions, such as riverbank filtration (Reungoat et al., 2011) and constructed wetlands (Lutterbeck et al., 2017), are increasingly recognized as promising alternatives for dealing with water purification, with minimal or no requirements for chemical inputs (Wang et al., 2018). Therefore, in order to evaluate the potential benefits of reduced chemical inputs during wastewater treatment processes, we assumed that there may be alternative nature-based solutions capable of achieving similar quality of reclaimed water to that delivered by the three primary scenarios (referred to as the Chemical-Free Solution). In the literature, the use of sludge nutrients for crops was found to reduce the life cycle impacts by restricting the production and application of commercial fertilizers (Wang et al., 2018). To this end, land use of the remaining nutrients was also considered in the following discussion (referred to as the Sludge Recycling Solution). The relevant assumptions and modelling methods for these three optimization strategies are provided in the Supplementary Information (Text S3). Hotspot analysis (Fig. 5) visualizes and isolates the key role of each of the above-mentioned solutions and their integration for the minimization of unwanted impacts generated in the three primary scenarios.

The major concerns with respect to the DSE scenario are associated with the life cycle eutrophication and non-carcinogenic human health impacts caused by elevated emissions of waterborne pollutants (particularly nutrients and HMs) from fertigation (Figs. 2a and 3b). An added implementation of energy recovery or chemical-free water purification

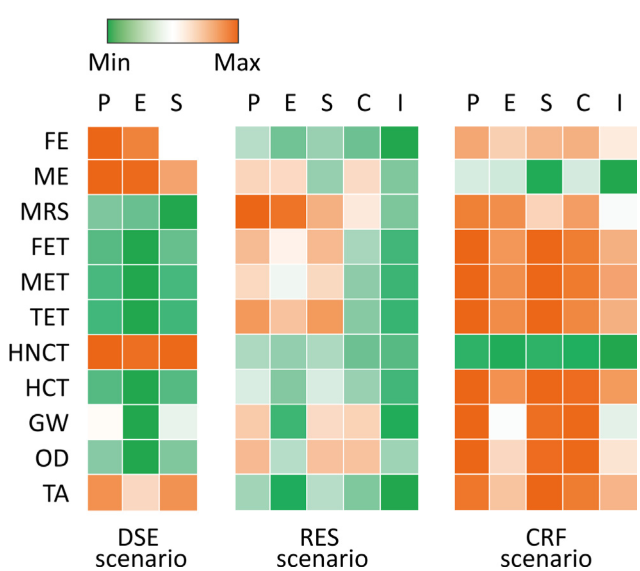

Fig. 5. Primary scenarios and optimization solutions with differentiated normalized impact hotspots. To obtain these normalized results (dimensionless) of the trajectories of environmental and human health impacts from each primary scenario with potential optimization solutions, the scores for each impact category, primary scenario, and alternative solution were normalized with the maximal and minimal results of the respective impact category. The color scale ranks the magnitude of the impacts (with orange denoting an important impact hotspot) and reveals the potential optimization (from orange to green). Abbreviations: FE, freshwater eutrophication; ME, marine eutrophication; MRS, mineral resource scarcity; FET, freshwater ecotoxicity; MET, marine ecotoxicity; TET, terrestrial ecotoxicity; HNCT, human non-carcinogenic toxicity; HCT, human carcinogenic toxicity; GW, global warming; OD, ozone depletion; TA, terrestrial acidification; P, primary scenario; E, Energy Recovery Solution; S, Sludge Recycling Solution; C, Chemical-Free Solution; and I, Integrated Solution. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

technologies was found to be somewhat beneficial with reductions in eutrophication, ecotoxicity, and/or additional unintended effects on the atmosphere. However, non-carcinogenic human health risks were still dominant (Fig. 5, Energy Recovery Solution and Chemical-Free Solution). This implies that the environmental and human health impacts of diverting secondary effluent to agriculture could be fully offset only if improvements in reclaimed water quality are prioritized.

The RES scenario, where implementation of chemical dosing to enrich the sludge phosphorus levels and land disposal of dewatered chemically enriched solids, produced relatively significant impacts on life-cycle mineral resource scarcity (Fig. 2c). However, there are clearly alternative sludge routes that could be considered, from unregulated dumping (with all the resulting environmental impacts), to regulated agriculture and application (with obvious nutrient use), and to high technology processing and energy extraction. As shown in Fig. 5 (Sludge Recycling Solution), diversion of chemical biosolids from landfill to land use significantly offset and mitigated the mineral resource depletion impact. This optimized performance is mainly attributed to the avoidance of production and use of commercial fertilizers. As the RES scenario was improved through energy recovery actions, its atmosphere-related impacts likely decreased, but not those of mineral scarcity. The findings of the Integrated Solution shown in Fig. 5 further demonstrate that integrating energy capture, biosolids application, and chemical-free water technologies with sophisticated wastewater reclamation processes can result in overall reductions in environmental and human health impacts, since low chemical-dependent water treatment processes likely provide complementary benefits of decreasing toxicity effects.

The CRF scenario involved tertiary configurations, such as energy-intensive RO, which enabled increased elimination of nutrients and toxic substances (HMs and CECs) from reclaimed water. The CRF scenario therefore performed better than the other two scenarios in terms of achieving minimal non-carcinogenic human health risks and lower eutrophication potentials (Figs. 3b and 2a). Nevertheless, indirect emissions from power plants and the release of contaminants through traditional sludge land disposal were the main driving forces of the heavier burdens in other environmental and human health impacts observed in the CRF scenario. This can be verified once the implementation of capturing and utilization of biogases (CH_4 and N_2O) is taken into consideration (Fig. 5, Energy Recovery Solution). Similarly, integrated solutions (energy recovery, nutrient recycling, and reduced chemicals) were also effective in reducing the overall negative impacts of the CRF scenario (Fig. 5, Integrated Solution).

4. Implications and conclusions

Improving water security while addressing agricultural sustainability are among the core Sustainable Development Goals under Agenda 2030 of the United Nations (UN, 2017b). Many countries and regions are devoting substantial efforts to transform the existing wastewater treatment infrastructure into resource recovery facilities and promoting end-use of recovered products in the agricultural sector (UN, 2017a). It has become evident that water reclamation for crop irrigation is a promising solution in regions with water shortages and provides benefits in reducing unintended impacts resulting from exploitation of traditional irrigation water resources (BCCR, 2017). Nonetheless, implementation approaches for water reclamation have not yet been standardized to any great extent, while regulations restricting emerging pollutants in reclaimed water are still in their infancy. Hence, insight into the life cycle burdens and benefits attributed to different water reclamation implementations becomes vital, but still remains unclear.

In this study, we found that there is no silver bullet for water resource recovery. As we design high-performance wastewater reclamation systems and repurpose reclaimed water for agricultural applications, it is important to recognize and consider the potential trade-offs of different wastewater purification implementations, so that decisions made prior to large scale practices are well informed. Our findings show that the net production of reclaimed water for agricultural irrigation are mainly dependent upon the sophistication of process configurations (in terms of multistage combinations of secondary processes or equipping with complex tertiary units), and that the direct benefits of water resource recovery could be offset by other adverse environmental and human health impacts. As such, it is necessary to perform explicit analysis on the system-level impacts and trade-offs during the planning and design stage decision-making. Our findings further indicate that these adverse environmental and human health impacts are dependent on energy and chemical inputs (such as iron chloride for enhanced phosphorus removal), which provide beneficial information for researchers and engineers to explore solutions at reducing the use of energy- or chemical-intensive processes.

While not explicitly investigated in the present study, results from an expanded system optimization suggest that concurrent implementation of energy recovery, nutrient recycling, and nature-based measures should be taken into consideration during implementation of water reclamation practices, particularly as water purification facilities include input-intensive treatment chains (Hao et al., 2019; Shiu et al., 2017). Furthermore, specific geographical, climate, or other considerations can help offset the impacts. For example, the electricity mix was found to significantly affect the overall impacts of energy-intensive water reclamation practices (Fig. S2). When a region is rich in renewable energy sources, such as wind, energy-intensive water purification technologies, such as RO, can be applied without significant energy-related and GHG impacts (Fig. S2). Therefore, future research efforts should necessarily be devoted to more holistic analyses of the trade-offs and synergies associated with the nexus of water, agriculture, and energy sectors for sustainable resource recovery processes, as well as on different local conditions.

The focus of this study was on the systematic evaluation of reclaiming water resources from different implementation approaches for irrigating crops to characterizing the application limits and pinpointing optimization strategies while relying on hybrid models. However, our analysis could be refined further once more reliable model inputs are made available. Here, the major source of uncertainty arose from the assumptions with respect to emerging contaminants in reclaimed water. In the context of mainstreaming LCA, impact assessment approaches still do not include CFs for many existing CECs, or the estimation parameters are significantly uncertain (Heijungs and Lenzen, 2014). In this analysis, we combined an additional 19 essential CECs obtained from the literature with existing CECs in the USES-LCA model to measure the ecotoxicity and human health impacts. Nevertheless, the total number of CECs considered in this study was still only 40% of the CECs currently identified in urban wastewater systems (Rahman et al., 2018), since their toxicity effects and removal mechanisms through water and wastewater treatment are not totally characterized or understood to date (Kim et al., 2018). In addition, human health implications were captured by the mid-point based LCA approach, but relied only on the toxicity effects of chemicals. A recent attempt to include microbial risks in the LCA of wastewater treatment management through blending quantitative microbial risk assessment (QMRA) highlights the potential of addressing the health effects of water reclamation from both the chemical and microbial risk perspectives (Harder et al., 2014, 2016). However, a detailed analysis incorporating microbial pollutants and their risks is beyond the scope of this study as the integration of LCA and QMRA is still in its infancy, with their implementation being significantly influenced by differences in their analytical components (Harder et al., 2015). Future exploration involving much more CECs and accounting for microbial hazards in reclaimed water will benefit a more comprehensive assessment and advance our understanding of the overall toxicity of water reclamation for non-potable or even potable uses, once the advances in relevant studies have been achieved. Additional model parameters with simplified assumptions in this study—such as rate of nitrogen leaching from sludge disposal on land, transport distance between water reclamation and end-use sites, and crops type—should be carefully investigated in future studies to better understand their potential influences under specific and real-life conditions.

Overall, this study provides essential information to a broad audience to create a synergic impetus for the development and implementation of timely and truly sustainable solutions to current water and agricultural challenges.

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.

Acknowledgement

We acknowledge support from the Beijing Talents Foundation, China (2017000021223ZK07), the Beijing Nova Program, China (Z171100001117078), the National Natural Science Foundation of China (51922013), and the Youth Innovation Promotion Association of the Chinese Academy of Sciences, China (2016041), and the Royal Society Newton International Fellowship, UK (NF160404).

Author contributions

X.W. conceived the research. Y.R.P. conducted the research. X.W. and Y.R.P. analyzed the data. X.W. and Y.R.P. wrote the paper. Z.J.R, C.Z.H., J.L., and D.B. contributed substantially by commenting on and revising the paper.

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2019.105266.

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