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DOI 10.1016/j.scitotenv.2023.166520

Publication date 2023 Document Version Final published version

Published in Science of the Total Environment

# Citation (APA)

Bhambhani, A., Kapelan, Z., & van der Hoek, J. P. (2023). A new approach to circularity assessment for a sustainable water sector: Accounting for environmental functional flows and losses. *Science of the Total Environment*, *903*, Article 166520. https://doi.org/10.1016/j.scitotenv.2023.166520

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# Science of the Total Environment

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



# A new approach to circularity assessment for a sustainable water sector: Accounting for environmental functional flows and losses



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# HIGHLIGHTS

# GRAPHICAL ABSTRACT

- Circular flows are redefined to include the biogeochemical resources.
- Returning biogeochemical resources to their natural cycle constitutes circularity.
- New assessment method shows higher circularity than the original MCI.
- Treated wastewater fertigation improves water and nitrogen circularity.





## ARTICLE INFO

Editor: Ashantha Goonetilleke

Keywords: Circularity assessment Biogeochemical resources Resource recovery Wastewater reuse Fertigation

# ABSTRACT

Resource recovery solutions can reduce the water sector's resource use intensity. With many such solutions being proposed, an assessment method for effective decision-making is needed. The water sector predominantly deals with biogeochemical resources (e.g., nitrogen) that are different from technical resources (e.g., industrial coagulants) in three ways: (1) they move through the environment in natural cycles; (2) they fulfil different human and environmental functions; and (3) they are subject to substantial environmental losses. Whilst several circularity assessment methods exist for technical resources, biogeochemical resources have received less attention. To address this, a well-established material circularity indicator (MCI) method is modified. This is done by redefining the terms: restoration, regeneration, and linear flows to create a new circularity assessment approach. The new approach is demonstrated in a real-life case study involving treated wastewater (TW) fertigation. The new approach reveals that using the original MCI method underestimates the circularity of resource recovery solutions involving biogeochemical resources. This is because, in the original MCI method, only the flows that are reused/recycled for human functions can be considered circular, whereas, in the new approach, one also considers flows such as N2 emission and groundwater infiltration as circular flows. Even though these may not be reuse/recycle type flows, they still contribute towards future resource availability and, thus, towards sustainability. The modified assessment method shows that TW fertigation can significantly improve nitrogen and water circularity. However, careful planning of the fertigation schedule is essential since increasing fertigation frequency leads to lower water but higher nitrogen circularity. Additionally, collecting drainage water for

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https://doi.org/10.1016/j.scitotenv.2023.166520

Received 15 May 2023; Received in revised form 24 July 2023; Accepted 21 August 2023 Available online 22 August 2023

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## 1. Introduction

Growing resource use intensity and waste production are causing scarcity of resources and environmental pollution. Therefore, reducing the reliance on virgin resources and avoiding their dissipation are fundamental for sustainability, and this is well understood in the water sector. Consequently, concepts of carbon neutrality (Mo and Zhang, 2012), wastewater reuse (Lyu et al., 2016), and nutrient recovery (Mo and Zhang, 2013) have been explored in recent years. The strategies to recover resources from the urban water cycle can be broadly classified as resource recovery solutions. As a part of the circular economy, they are meant to decouple economic development from resource extraction by recirculating resources (Corona et al., 2019).

Different resource recovery strategies can contribute to resource conservation to varying degrees. Therefore assessment methods are needed to select the most effective CE transition strategy (de Oliveira et al., 2021) and to measure progress towards the circular economy (Saidani et al., 2019). However, circularity assessment in the water sector can get complex because of a mix of technical (e.g., industrial coagulants) and biogeochemical (e.g., nitrogen) resources. The technical resources are generally abiotic, non-renewable, and synthetic and have the potential to remain circulating in the production system (i.e., industrial manufacturing, recovery, and reuse) (Braungart et al., 2007; Ellen MacArthur, 2015; Mestre and Cooper, 2017), without being disposed in landfills or used as fuel for energy generation (Navare et al., 2021). Biogeochemical resources move in a continuous cycle, passing alternatively between a non-living form and as part of living matter (Bertrand et al., 2015). While most assessment methods are designed for the technical cycle resources, not much research has gone into the circularity assessment of biogeochemical resources.

The circularity assessment of biogeochemical resources is not straightforward because of three factors. Firstly, these resources (e.g., water) naturally recirculate (e.g., in the hydrological cycle); hence they can become scarce because a particular resource form may not be easily used (e.g., water vapour) or accumulate in an environmentally pernicious form (Rijsberman, 2006) (e.g., untreated wastewater). Secondly, while flowing through the cycles, different forms of biogeochemical resources serve different environmental functions (Gleeson et al., 2020; Zipper et al., 2020) (e.g., while the water flowing in an over-land stream sustains aquatic ecosystems, evaporating water helps to cool down the environment). Therefore, simply maximising a particular resource form for human benefits can disrupt critical ecosystem functions. Finally, the availability of biogeochemical resources can be significantly affected by the environmental loss mechanisms (Vicente-Serrano et al., 2014) (e.g., evaporation loss of irrigation water). These losses have to be determined according to local conditions; otherwise, the circularity assessment remains superficial. Further complications arise because some of these losses on a smaller spatial and temporal scale may be beneficial on a larger scale (Grafton et al., 2018). Thus, certain biogeochemical flows categorized as losses can also be considered circular because they enable future resource availability. Guidance is lacking on how to determine if a certain resource flow can be categorized as linear or circular.

Given above, what constitutes circular flows is different for biogeochemical resources than for technical ones, and a different approach to assessing circularity is needed. Current methods designed for technical resources may underestimate or overestimate the circularity of resource recovery solutions if applied to biogeochemical resources. For the circular economy to support sustainability, the definition of the circularity of all resources should be aligned with sustainability.

This study aims to develop a new and improved approach to the circularity assessment of biogeochemical resources commonly found in

the water sector in two steps. Firstly, current assessment methods do not account for environmental functional flows. In this paper, a method is introduced for segregating biogeochemical flows as linear or circular that ensures that the resource flows serving environmental functions are counted towards improved circularity. Also, very few studies base their circularity assessment on resource flow models accounting for the local conditions. Here, a more realistic circularity assessment of biogeochemical resources is achieved by basing the assessment on a resource flow model that accounts for the local climate and resource application schedule.

Section 2.1 starts with the definitions of circularity and sustainability, and it highlights that circularity should be assessed in a way that can support sustainability. Next, some circularity assessment methods are discussed in Section 2.2 to indicate that they are mainly suited for the technical cycle resources. Thereafter in Section 2.3, three factors related to the biogeochemical resources are presented that make defining and assessing their circularity more complicated than doing so for the technical resources. In Section 2.4, the existing material circularity indicator (MCI) method is described, the restorative, regenerative, and linear flows, as originally introduced by the Ellen MacArthur Foundation (2019) as part of their MCI method, are presented, and also the lack of consensus on how to define these terms is discussed. Next, new definitions for the three terms (linear, restorative, and regenerative flows) and the new assessment approach based on the modification of the MCI method are presented. The new circularity assessment approach is demonstrated in a real-life case study involving treated wastewater fertigation in Section 2.5. Also, DSSAT and CLIMWAT, tools for modelling resource flows, are briefly discussed in this section. Section 3 presents the circularity assessment results. This is followed by a discussion of the case study results in Section 4, where factors that improve water and nitrogen circularity in fertigation are analysed. Subsequently, the differences between the new assessment approach and the original MCI are presented. Finally, the conclusions about the new circularity assessment method and the fertigation case study follow in Section 5.

# 2. Material and methods

# 2.1. Circularity and sustainability

The concepts of sustainability and circularity are introduced here, not to discuss the details of their various definitions. The purpose is to support the opinion of the authors that circularity should be defined and assessed in a way that supports sustainability. This is to avoid ignoring the wider environmental implications of the resource recovery solutions and thereby propagating circularity for circularity's sake (Harris et al., 2021).

Several definitions of sustainability exist, but the most popular one is based on Brundtland (1987): Economic development that meets current needs without compromising the needs of future generations. Conserving natural resources for future generations is thus essential for sustainability, and this is where the circular economy fits in.

The circular economy is a concept of recirculating resources within the economic system to maximise the value recovered from them. This concept has been developed as an alternative to the linear economy, where resources are extracted, used, and discarded as waste (Corona et al., 2019). The goal of resource recovery solutions is to decouple economic development from virgin resource extraction and thereby promote sustainability (Bhambhani et al., 2022). Although circularity is meant to promote sustainability (Corona et al., 2019; Terra dos Santos et al., 2022), it may not always do so (Mancini and Raggi, 2021; Terra et al., 2022). Thus, it is crucial to define and assess circularity in a way

# that supports sustainability (Harris et al., 2021).

## 2.2. Circularity assessment

Since a method is needed to assess the water sector's circularity transition, some of the current methods were analysed. Since most methods are focussed on the technical cycle (Navare et al., 2021; Rocchi et al., 2021), these are discussed first. Collection rates (Haupt and Hellweg, 2019), the percentage of a resource collected after use for recycling, may be used to measure circularity. The recycling rate (Haupt and Hellweg, 2019) is another similar indicator representing the fraction of resources that becomes part of a secondary product. Thus, higher collection and recycling rates may imply higher circularity. In the case of using treated wastewater for irrigation, even though avoiding freshwater will lead to improved circularity, only a tiny percentage of the irrigation water becomes part of the crop (secondary product). A large portion is evaporated, transpired, or seeps underground, leading to a low recycling rate yet contributing to groundwater recharge and thus water sustainability (Kazem Attar et al., 2020). The circular economy index method (Di Maio and Rem, 2015) assesses circularity as the ratio between the market economic value produced by a recycler to the material economic value entering a recycling facility. This is fine from an economic point of view, but a method solely based on economic value maximisation might lead to biogeochemical resources being diverted towards activities that generate the highest economic returns even at the cost of maintaining environmental functions (e.g., improving irrigation efficiency at the cost of groundwater recharge). The circularity indicator developed by Franklin-Johnson et al. (2016) measures the time duration of resource use, focussing on 'materials moving perpetually within industrial systems' (Franklin-Johnson et al., 2016). This indicator maximises resource access for human functions which can lead to undesirable consequences. This is demonstrated by the fact that maximising treated wastewater reuse at the cost of reduced discharge into a stream is known to cause a reduction of stream flow quantity and degradation of the stream water quality (Wolfand et al., 2022).

Next, the methods directly relevant to the water sector are discussed. While several water balance studies exist, including Kenway et al. (2011), Venkatesh et al. (2017), and Currie et al. (2017), circularity assessment of water and the resources present in water has received very little attention (Arora et al., 2022; Renfrew et al., 2022). Preisner et al. (2022) compiled a set of indicators for the circularity assessment of the water sector. They proposed a method using the average of the recovery rates of nutrients, and organic matter, the reuse rate of treated wastewater, and the energy sufficiency of a WWTP. This indicator is relatively simple to calculate and can help summarize the WWTP performance in recovering important resources. But, the application scope of this indicator is limited to a WWTP and does not include the resource application process (e.g., irrigation). Kakwani and Kalbar (2022) have developed the water circularity indicator (WCI) based on the MCI method developed by the Ellen MacArthur Foundation (2019). A city-wide urban water circularity framework has been developed by Arora et al. (2022). Both approaches help to assess the urban water systems but exclude hydrological flows such as evaporation, transpiration, runoff, and infiltration losses. Nearly 70 % of the total water used by humans is for agricultural irrigation (Cassardo and Jones, 2011), and flows such as evapotranspiration, runoff, and infiltration constitute a large part of the agricultural water flows (Kazem Attar et al., 2020). Therefore, a discussion needs to be started about modelling these environmental losses and assessing their effect on circularity.

Based on the discussion above, three observations about the current assessment methods are presented. Firstly, with these methods, one assumes that retaining resources for human use alone (through high collection rates, high recycling rates, and lengthening the use duration) constitutes circularity, i.e., resource availability for human functions is maximized while every other flow is considered as a 'waste'. This may be appropriate for technical resources for which defining 'waste' flows such as landfills is straightforward. However, for the biogeochemical resources, 'waste' flows may also be beneficial as long as they contribute towards the sustainability of a resource. For example, irrigation water leakage can contribute to a large portion of groundwater recharge in some regions (Bouimouass et al., 2020; Qi et al., 2023). Secondly, using the existing methods that maximise the human functions of resources can backfire and lead to resource scarcity. To illustrate, maximising agricultural water efficiency by reducing groundwater infiltration is known to contribute to groundwater scarcity and reduce environmental flows necessary for sustaining aquatic ecosystems (Batchelor et al., 2014; Simons et al., 2020). Thirdly, when using the existing assessment methods, one cannot account for the environmental losses of biogeochemical resources, a factor that substantially affects circularity. For example, nitrogen recovered from wastewater can be applied to agricultural soil as a recycled fertilizer. However, the recycled nitrogen that leaches into groundwater cannot be considered circular because this will contaminate the groundwater. And the amount of nitrogen leaching strongly depends on factors such as climate, precipitation (Jabloun et al., 2015), and application rate (Bowles et al., 2018; Shepherd, 1996). Therefore, modelling these factors is crucial for an accurate assessment.

Therefore, the discussed methods do not take into account the complexities of assessing the circularity of biogeochemical resources.

# 2.3. Biogeochemical and technical resources

Resources can be categorized as technical and biological (Ellen MacArthur, 2015; Moreno et al., 2016). The technical resources are generally abiotic, non-renewable, and synthetic and have the potential to remain circulating in the technical cycle (i.e., industrial manufacturing, recovery, and reuse) (Braungart et al., 2007; Ellen MacArthur, 2015; Mestre and Cooper, 2017), without being disposed in landfills or used as fuel for energy generation (Navare et al., 2021). For example, metals need to be reused as many times as possible to avoid their disposal in nature (Velenturf et al., 2019). In contrast, biological resources can safely cycle between the technical cycle and the natural environment (Braungart et al., 2007; Moreno et al., 2016).

'Biogeochemical' is a better term to describe those resources that can cycle between the technosphere and the natural environment since these resources need not always be of biological origin. Biogeochemical resources are those resources that move in a continuous cycle, passing alternatively between a non-living form and as part of living matter (Bertrand et al., 2015). This study is concerned with the short-term biogeochemical cycles with a time scale of a few years, such as the short-term carbon or nitrogen cycle. For the same reason, phosphorus is not included because it is mainly mined from non-renewable phosphate rocks (Liu et al., 2023; Scholz et al., 2013).

There are three main differences between the two resource categories that are relevant to circularity. Firstly, differentiating between the linear and circular flows for biogeochemical resources is more complex. Landfilling or energy recovery are linear flows for technical resources because they render these resources permanently unavailable for future use. In contrast, biogeochemical resources naturally flow in short-term cycles (i.e., several years), making it more complicated to differentiate between their linear and circular flows. For example, releasing inert N<sub>2</sub> into the atmosphere can be considered a waste flow (van der Hoek et al., 2018), but simultaneously, closing the nitrogen cycle (by releasing N<sub>2</sub> back into the atmosphere) is considered a pathway for indirect reuse of nitrogen (Spiller et al., 2022) and a way to remove excess reactive nitrogen from the environment (Galloway et al., 2021). So, are dinitrogen emissions from the WWTP linear or circular flows?

Secondly, unlike technical cycle resources, biogeochemical resources serve both human and environmental functions. For instance, treated wastewater (TW) discharge is beneficial for sustaining stream flows and the ecosystems linked to it (Rice and Westerho, 2017) (environmental function), but the TW may be diverted for irrigation (human function), leading to a stream flow reduction and potential ecosystem losses (Rice and Westerho, 2017). So does complete water circularity mean maximising water use for human benefits at the cost of the ecosystems?

Thirdly, environmental loss mechanisms can be a significant cause of biogeochemical resource scarcity, despite recycling. For example, recovered nitrogen used in agriculture may be dissipated in reactive forms causing substantial economic and environmental damage (Müller and Clough, 2014). Similarly, rising evaporative water loss can be a cause of water scarcity (Vicente-Serrano et al., 2014). Such environmental losses depend on the local climate, soil characteristics, rainfall, etc. Furthermore, some losses may even contribute to circularity. For instance, groundwater infiltration is considered to be irrigation loss at the farm scale, but this 'loss' may simultaneously recharge groundwater (Grafton et al., 2018; Kazem Attar et al., 2020). So, should infiltration loss of water be considered a linear flow even though it may prevent future water scarcity?

To assess the circularity of biogeochemical resources in a way that supports sustainability, the complexities of the biogeochemical cycles have to be considered. Fig. 1 shows a typical technical resource cycle and the nitrogen cycle as an example of a biogeochemical resource to illustrate how much more complicated the flows of the latter can be.

# 2.4. The material circularity indicator

Out of the circularity assessment methods discussed in Section 2.2, the MCI is the most promising for the water sector because of two reasons. Firstly, the MCI covers the input as well as the output circularity of a process. Therefore, one can assess the percentage of the resource feedstock as well as products that can be considered circular. Secondly, the MCI relies on readily-available mass or volume data. Thus, as also considered by Kakwani and Kalbar (2022), the material circularity indicator (MCI) provides a good starting point for developing a novel circularity assessment method for the water sector. Still, it is only a starting point as the MCI method, too has limitations to be addressed.

The MCI method is based on differentiating restorative and regenerative flows from linear flows. Restorative flows are defined as those that are reused/recycled, and linear flows are the ones that originate from virgin sources, ending up in landfills or energy recovery processes (Ellen MacArthur Foundation, 2019). Regeneration refers to the returning of biotic resources to the natural environment such that the resources remain biologically accessible and the production capacity of the natural source is maintained (Ellen MacArthur Foundation, 2019). However, these definitions do not help distinguish between linear and restorative/regenerative biogeochemical flows. For example, recovered nitrogen applied to the soil may leach into groundwater which is a linear flow even though it is not landfilled nor used for energy recovery. Thus, direct application of the MCI method to the water sector resource recovery solutions is problematic because this restricted way of defining linear and restorative/regenerative flows does not apply to most biogeochemical resources.

#### 2.4.1. Original MCI method

Below, the first two steps of the MCI method are shown to calculate the virgin resource input and the unrecovered waste of a process, and then these steps are used as a framework for calculating biogeochemical resource circularity.

## 1. Calculate virgin resource input as follows:

$$V = M(1 - F_R - F_U - F_S)$$
(1)

where V is the virgin resource input, M is the total resource input,  $F_R$  is the feedstock fraction derived from recycled sources,  $F_U$  is the feedstock fraction derived from reused sources, and  $F_S$  is the fraction of biological resources obtained from sustained production.

2. Calculate the unrecovered waste output as follows:

$$W = M(1 - C_R - C_U - C_C - C_E)$$
<sup>(2)</sup>

where M is the total resource input,  $C_R$  is the fraction of the resource flowing into a recycling process,  $C_U$  is the fraction of the resource flowing into component reuse processes after the use phase,  $C_C$  is the fraction composted, and  $C_E$  is the fraction originating from sustained biological production and used for energy recovery.

As can be seen from Eqs. (1) and (2), the MCI requires the estimation of linear flows by subtracting the regenerative/restorative flows from the total resource throughput. Resources that originate from reuse/ recycle sources or from sustained biological production are considered restorative/regenerative. On the other hand, all resource outputs that go into a reuse/recycle process, are composted, or are used for energy recovery are considered restorative/regenerative. These definitions of regenerative/restorative flows work well for technical resources but not for biogeochemical flows. For example, WWTPs denitrify nitrogen oxides to emit nitrogen gas (N<sub>2</sub>) and nitrous oxide (N<sub>2</sub>O). Should inert N<sub>2</sub> emissions be considered linear only because they are not reused/recycled? Clear guidance is lacking that would help to classify biogeochemical flows as regenerative or linear.

# 2.4.2. Redefining restorative, regenerative, and linear flows

Morseletto (2020) has pointed to the lack of a clear definition for the



Fig. 1. (a) A typical technical resource cycle for which it is relatively simple to differentiate between circular (green) and linear flows (red); (b) The nitrogen cycle makes it complex to segregate circular and linear nitrogen flows.

terms 'restoration' and 'regeneration' in CE literature and has defined the term 'restoration' as a return to a previous or original state. Morseletto (2020) also proposes a definition for 'regeneration' as aiding the self-renewal capacity of natural systems against overexploitation by humans. But, the authors also suggest omitting the concept of 'regeneration' as a central CE principle due to a lack of robust guidance on how to apply this concept to resource recovery solutions. Since returning a resource to a previous state for human use and releasing the resource into nature are two very different kinds of flows, the term 'regeneration' is retained here as a CE principle. The terms restorative, regenerative and linear flows are redefined as follows:

Restorative flow is that which recovers a resource for direct human use (e. g., recovery of the struvite fertilizer out of wastewater through precipitation).

Regenerative flow is that which returns a resource to the state in which it was originally appropriated from nature for human use. This is to promote the self-renewal and ecosystem-sustaining capacity of biogeochemical cycles in response to overexploitation (e.g., releasing reactive nitrogen as  $N_2$  into the atmosphere to close the nitrogen cycle).

Linear flow is that which is obtained from virgin sources and/or discarded in a form different from how the resource was originally obtained for human use (e.g., returning water obtained from a river as water vapour to the atmosphere).

These definitions can now be used to differentiate between restorative, regenerative, and linear biogeochemical flows. For example, nitrogen (N<sub>2</sub>) is converted into biologically active forms through the Haber Bosch process (Razon, 2018), and if this nitrogen is returned to the atmosphere as N2, then the return flow can be considered regenerative. On the contrary, water obtained from a freshwater source in liquid form, used for irrigation and returned to nature as water vapour is a linear flow. While it is true, that any water body exposed to the atmosphere will have some evaporation but this is usually a natural process and of a much smaller magnitude compared to the evaporation from an irrigated field. A concern may be raised about the high energy use of obtaining reactive nitrogen from the atmosphere. Energy use is an inevitable factor to be considered for the sustainability of a process. However, the concept of energy use should not be mixed up with circularity. Often high circularity comes at the cost of high energy (Campbell-Johnston et al., 2019; Gregson et al., 2015). Even certain nitrogen recovery technologies, such as air stripping, can have an energy consumption in the same order as required for fixing atmospheric nitrogen using the Haber Bosch process and converting back to N2 using nitrification-denitrification (van der Hoek et al., 2018).

# 2.4.3. New material circularity assessment approach

The circularities of the biogeochemical and technical resources are assessed using the following equations:

1. Calculate the virgin inputs as follows:

V = M(1 - RSIF)(3)

where V is the virgin resource input, M is the total resource input, and RSIF is the restorative input fraction comprised of the input resource that originates from the same or another use process.

# 2. Calculate the unrecovered waste output as follows:

$$W = M(1 - RSOF - RGOF)$$
<sup>(4)</sup>

where W is the total unrecoverable waste, M is the total resource input, RSOF is the restorative output fraction defined as accumulated resource fraction (below saturation) or modified to a previous state for human functions, and RGOF is the regenerative output fraction defined as the resource fraction modified to the original state in which these were obtained from nature.

3. Calculate the linear flow indicator as follows:

$$LFI = \frac{V + W}{2M} \tag{5}$$

where LFI is the linear flow indicator, V is the virgin resource input (Eq. (3)), W is the unrecoverable waste output (Eq. (4)), and M is the total resource input.

4. Calculate the material circularity indicator as follows:

$$MCI = (1 - LFI) \times 100 \tag{6}$$

where MCI (%) is the material circularity indicator of a resource, and LFI is the linear flow indicator of the resource (Eq. (5)).

In the original MCI method, a utility factor (F(X)) is used to penalize for potentially lower product durability resulting from recycled ingredients. However, the industrial analogy of product durability does not apply to the water sector (Kakwani and Kalbar, 2022), and hence, the utility factor is excluded from the MCI calculation. In the next section, the new circularity assessment method is demonstrated in a case study. The new MCI values are generated and compared to the values of the original MCI method.

# 2.5. Case study

### 2.5.1. System description

A block scheme of the Corleone case study in Italy is shown in Fig. 2. An activated sludge wastewater treatment plant (WWTP) treats 3700  $m^3/d$  of domestic wastewater (Mannina et al., 2022). A portion of the treated wastewater (TW) will pass through an ultrafiltration unit which is added to the existing WWTP to produce irrigation water for agriculture (Mannina et al., 2022). The system boundary starts at the point from where the irrigation water is sourced for irrigation and ends at the agricultural land. For assessing a resource recovery solution, the WWTP should also be included within the system boundary. But, in this demonstration, the choice of a narrower system boundary is inspired by two reasons. Firstly, the difference in the assessment results between the original MCI and the modified approach will show up in the irrigation process because of environmental losses such as groundwater infiltration. Such losses do not form any significant part of the flows through the WWTP, and thus, the difference in the assessment results will not be substantial. Secondly, since the focus is on the demonstration of the new method, including only the irrigation process allows for simplicity without losing any generalizability. The circularity is assessed over the crop growing period within a year.

# 2.5.2. Water and nitrogen circularity assessment

First, the circularity of irrigating every three days using freshwater (FW 3) is compared to using treated wastewater (TW 3). Next, the circularities of fertigation every three (TW\_3) and ten days (TW\_10) are compared since the schedule is an important factor affecting water and nutrient balance in agriculture (Mermoud et al., 2005). The recommended irrigation schedule for the tomato crop is highly locationdependent and can range from every three days (Shao et al., 2010) to every ten days (Karuku et al., 2014). Since no irrigation schedule was specified by the case study owners, a three-day and a ten-day schedule are used to cover a wide range. Furthermore, the most extensively used soil and water management intervention in agriculture is subsurface or tile drainage (TD) (Williams et al., 2015). Tile drains are pipes installed underground to collect percolating irrigation water and enable drainage water recycling (DWR) (Ghirardini and Verlicchi, 2019), which is the practice of collecting drained water from fields in a reservoir for use in times of soil water deficit (Reinhart et al., 2019). Tile drainage collection also helps to reduce nutrient load to water reservoirs by preventing the



Fig. 2. Block scheme for reuse of treated wastewater (TW) for fertigation in Corleone, Italy. The circularity for the irrigation process is assessed.

discharge of nutrient-rich irrigation water (Reinhart et al., 2019). The effect of drainage water recycling (DWR) on the water and nitrogen circularity is analysed.

Therefore, the circularity results from the original MCI and the modified approach are compared for four alternatives: (1) Irrigation using freshwater from the river and industrial nitrogen fertilizer application every three days (FW\_3); (2) Fertigation using treated wastewater every three days (TW\_3); (3) Fertigation using treated wastewater every ten days (TW\_10); (4) Fertigation using treated wastewater every ten days with drainage water recycling (TW\_10\_DWR).

# 2.5.3. Water and nitrogen supply

To assess the water circularity of this resource recovery solution, the irrigation water quantity is required, which depends on the choice of the crop, the climate, and the irrigation method. For the assessment, a field area of 200,000 m<sup>2</sup> (20 ha) is assumed, and the method of irrigation is drip irrigation with an irrigation efficiency of 85 %. An irrigation efficiency (IE) of 100 % means that all the irrigation water supplied is used either for a crop's evapotranspiration (ET) or stored in the soil for future use (Malik and Dechmi, 2019). Thus, an 85 % IE implies that 15 % of the supplied water is neither part of ET nor stored in the soil. This water is assumed to be evaporated during the water application. The tomato crop is irrigated using treated wastewater which requires 400–600 mm of water over its growing season of 90–150 days (FAO, 2022b), which translates to 80,000–120,000 m<sup>3</sup> for a 200,000 m<sup>2</sup> field. The growing period is 108 days, from 1 June 2021 to 16 September 2021.

The climate data for the nearest (Palermo) weather station was obtained using the CLIMWAT tool (FAO, 2022a). CLIMWAT is a climate database that enables the calculation of crop water requirements, irrigation supply, and scheduling based on climate data across the globe. Based on the temperature data from the Palermo weather station gathered using the CLIMWAT tool and the crop coefficient obtained from FAO (2022b), shown in Fig. 3, the ET requirement for tomatoes is estimated over the growing season to be in the order of  $112.1 \times 10^3$  m<sup>3</sup>. Adding up monthly rainfall data obtained using CLIMWAT, total rainfall in the order of  $10.6 \times 10^3$  m<sup>3</sup> is obtained as the difference between ET requirement and rainfall. Assuming an 85 % irrigation efficiency gives  $119.4 \times 10^3$  m<sup>3</sup> of gross irrigation water estimation, as shown in Table 1. The complete calculations are shown in the supplementary material.

Since TW nitrogen can serve as a secondary source of fertilizers, the nitrogen circularity is also calculated. The total nitrogen concentration in the Corleone effluent is 20 mg N/L (or 0.02 kg N/m<sup>3</sup> water), which lies within the concentration range of 5 and 30 mg N/L specified by Chojnacka et al. (2020). With a total fertigation water requirement of  $119.4 \times 10^3$  m<sup>3</sup> (see Table 1), this means a total of  $2.4 \times 10^3$  kg nitrogen is applied to the 20 ha field over the growing season. This is equivalent to 120 kg N/ha which falls within the 100 to 150 kg N/ha range of nitrogen requirement for tomato crops as specified by FAO (2022a).

Next, the three and ten-day irrigation schedules are entered into the



**Fig. 3.** Tomato crop evapotranspiration coefficient (Kc) for the different crop development stages (initial, mid, and end). Values obtained and figure adapted from FAO (2022b).

# Table 1

Irrigation water requirement for a 20 ha tomato field in Corleone, Italy, based on ET requirement and rainfall over the entire crop growing season from 01.06.21 to 16.09.21.

Month	Days	ET (x10 <sup>3</sup> m <sup>3</sup> )	Rainfall (x10 <sup>3</sup> m <sup>3</sup> )	Net irrigation requirement (x10 <sup>3</sup> m <sup>3</sup> )	Gross irrigation requirement (x10 <sup>3</sup> m <sup>3</sup> )
June	30	18.2	1.8	16.4	19.3
July	31	40.8	0.6	40.2	47.3
August	31	40.1	3.7	36.4	42.8
September	16	13	4.5	8.5	10
Total	108	112.1	10.6	101.5	119.4

Decision Support System for Agrotechnology Transfer (DSSAT) tool. DSSAT is a tool developed by an international network of scientists to integrate the knowledge of soil, crops, climate, and management for better decision-making in agriculture (Jones et al., 2003). DSSAT is used to model the fate of irrigation water and the nitrogen used for the fertilization of crops. The soil type is specified as deep sandy loam, and the field size is specified as 200,000 m<sup>2</sup> into DSSAT. The tomato crop and Sunny S—D 2010 cultivar are selected. The initial soil water content on the day of planting (01.06.21) was 106 m<sup>3</sup>. Based on the irrigation schedule and the precipitation, DSSAT calculates the soil water content on harvest day. The difference between the soil water on harvest and planting days has been specified as soil water used. In the next section, the obtained circularity results of the original and the modified MCI methods are presented.

# 3. Results

## 3.1. Nitrogen and water fate

DSSAT was used for simulating irrigation water and nitrogen fate for the four alternatives discussed in Section 2.5.2 using the inputs provided in Section 2.5.3. DSSAT calculates the soil water and nitrogen balance components. The soil water balance is shown in Table 2. The nitrogen balance can be found in the supplementary material.

The irrigation water inflow and the precipitation were specified by the authors, and DSSAT estimated the soil water used, which differs for the two irrigation schedules. The soil water used (difference between soil water content on the planting and the harvest day) is higher in the case of TW\_3 ( $17.1 \times 10^3$  m<sup>3</sup>) than for TW\_10 ( $13.1 \times 10^3$  m<sup>3</sup>). This may be due to the crop using more soil water in the three-day interval case due to higher evaporative losses. Further, the groundwater infiltration is higher for the TW\_10 ( $18.6 \times 10^3$  m<sup>3</sup>) than for the TW\_3 ( $6.8 \times 10^3$  m<sup>3</sup>) case. In the TW\_10\_DWR case, part of the drainage water ( $11.9 \times 10^3$  m<sup>3</sup>) is collected for reuse, while in the rest of the cases, all of the drainage water is assumed to recharge groundwater. While the soil evaporation is higher for the three-day interval ( $89.5 \times 10^3$  m<sup>3</sup>) than the ten-day interval irrigation ( $69.5 \times 10^3$  m<sup>3</sup>), the transpiration and crop uptake are higher for the ten-day interval case ( $3.7 \times 10^3$  m<sup>3</sup>).

The industrial/wastewater nitrogen input was specified, and DSSAT calculated the soil nitrogen used and mineralized nitrogen. Soil nitrogen used for the TW\_10 and TW\_10\_DWR alternatives was higher  $(0.3 \times 10^3 \text{ kg N/GS})$  than for the TW\_3 alternative  $(0.2 \times 10^3 \text{ kg N/GS})$ . This may be because, in the lower frequency applications, the crop relies more heavily on internal nitrogen cycling (Dawson et al., 2008). Nitrogen loss with drainage was higher for the TW\_10 case  $(0.7 \times 10^3 \text{ kg N/GS})$  than for the TW\_3 case  $(0.4 \times 10^3 \text{ kg N/GS})$  because more water infiltrates with lower frequency fertigation, also draining the nitrogen along with it. TW\_3 had a higher crop uptake of nitrogen  $(3.6 \times 10^3 \text{ kg N/GS})$  as compared to the TW\_10 and TW\_10\_DWR alternatives  $(3.5 \times 10^3 \text{ kg N/GS})$  decause high-frequency fertigation leads to higher crop uptake of nitrogen (Farneselli et al., 2015).

## 3.2. Circularity assessment

The original MCI method was applied to the above case study. For the FW\_3 alternative, both the water and nitrogen circularity values are 0. This is because freshwater was used for irrigation along with industrial nitrogen fertilizer, and on the output side, none of the nitrogen or water flows can be considered circular. The water and nitrogen

#### Table 2

Water inflows and outflows from DSSAT for the irrigation and rainwater specified in our case study. The water quantities are expressed in  $m^3/GS$  where GS stands for the growing season of the tomato crop running from 1 June to 16 Sept.

	FW_3 (x10 <sup>3</sup> m <sup>3</sup> / GS)	TW_3 (x10 <sup>3</sup> m <sup>3</sup> / GS)	TW_10 (x10 <sup>3</sup> m <sup>3</sup> / GS)	TW_10_DWR (x10 <sup>3</sup> m <sup>3</sup> / GS)
Inflows				
Treated wastewater input	0	119.4	119.4	119.4
Freshwater input	119.4	0	0	0
Precipitation	10.6	10.6	10.6	10.6
Soil water used	17.1	17.1	13.1	13.1
Outflows				
Irrigation loss	17.9	17.9	17.9	17.9
Groundwater infiltration	6.8	6.8	18.6	6.8
Drainage collected	0	0	0	11.9
Soil evaporation	89.5	89.5	69.5	69.5
Transpiration	29.5	29.5	33.3	33.3
Crop uptake	3.3	3.3	3.7	3.7

circularities for the TW\_3 alternative are 41 % and 29 %, respectively. This improvement (relative to FW\_3) is due to the use of treated wastewater containing nitrogen. For the TW\_10 case, water circularity improves further to 42 %. This slight improvement results from lower net soil water used in the ten-day interval than in the three-day interval irrigation schedule. The nitrogen circularity remains at 29 % for TW\_10. Finally, the water circularity of 46 % and nitrogen circularity of 37 % is obtained in the TW\_10\_DWR alternative. The higher water and nitrogen circularity values obtained in this alternative (compared to the TW\_10 alternative) are due to the drainage water collected for reuse.

The new MCI method was applied to the same case study. The water and nitrogen flows were classified as linear, restorative or regenerative flow. Fig. 4 shows the difference between regenerative and linear water flows as part of the original and the modified MCI. Precipitation, FW irrigation, and soil water use do not originate from reuse/recycle sources and, thus, are linear flows. Evaporation and transpiration are linear flows because the water used in liquid form is returned to the atmosphere as vapour. It was assumed that the water contained in the tomato would re-enter the WWTP through the human diet. Also, it was assumed that the crop residue is mulched and its water content replenishes soil water. Therefore, the crop uptake of water was treated as restorative. Irrigation water infiltration can contribute to groundwater recharge (Jia et al., 2020), and thus, infiltration was considered to be a regenerative flow.

Regarding nitrogen flows, soil nitrogen use, industrial nitrogen fertilizer addition, and soil nitrogen mineralization are considered linear input flows since they do not originate from any reuse/recycle sources. But, nitrogen added with treated wastewater is a restorative flow. Further, losses in the form of ammonia (NH<sub>3</sub>) or nitrogen oxide (NO) are both linear output flows because they are not reused/recycled. However, N<sub>2</sub> loss is a form of regenerative flow because nitrogen is released in the form in which it was originally obtained from the atmosphere.

To illustrate the new MCI method, the water MCI calculation steps for the TW\_3 alternative are shown here.

1. Calculate total water and nitrogen inflows as follows:

$$M_{water} = Gross \ irrigation \ water + Precipitation + Reduction \ in \ soil \ water$$
  
=  $(119.4 + 10.6 + 17.1) \times 10^3 = 147.1 \times 10^3 \ m^3$ 

- $M_N = Fertigation N + Reduction in soil N + Mineralized N$  $= (2.4 + 0.2 + 1.5) \times 10^3 = 4.1 \times 10^3 kg$
- 2. Calculate virgin inflows for water and nitrogen as follows: V = M(1 - RSIF)

$$V_{water} = \left(147.1\left(1 - \frac{119.4}{147.1}\right)\right) \times 10^3 = 27.7 \times 10^3 m^2$$
$$V_N = \left(4.1\left(1 - \frac{2.4}{4.1}\right)\right) \times 10^3 = 1.7 \times 10^3 kg$$

3. Calculate unrecovered water and nitrogen outflows as follows: W = M(1 - RSOF - RGOF)



Fig. 4. (a) Restorative/regenerative (green) and linear (red) water flows based on the original MCI. (b) Regenerative (green) and linear (red) flows based on the modified MCI. In the modified approach, flows such as crop uptake and groundwater infiltration of water are counted as restorative/regenerative flows unlike in the original MCI approach.

$$W_{water} = \left(147.1\left(1 - \frac{6.8}{147.1} - \frac{3.3}{147.1}\right)\right) \times 10^3 = 137 \times 10^3 m$$
$$W_N = \left(4.1\left(1 - \frac{3.6}{4.1} - \frac{0}{4.1}\right)\right) \times 10^3 = 0.5 \times 10^3 kg$$

4. Calculate the linear flow indicators as follows:

$$LFI = \frac{V + W}{2M}$$

$$LFI_{water} = \frac{27.7 + 137}{2 \times 147.1} = 0.56$$

$$LFI_N = \frac{1.7 + 0.5}{2 \times 4.1} = 0.27$$

5. Calculate the material circularity indicators as follows

 $MCI = (1 - LFI) \times 100$ 

$$MCI_{water} = (1 - 0.56) \times 100 = 44\%$$

$$MCI_N = (1 - 0.27) \times 100 = 73\%$$

Table 3 shows the results of the modified MCI and the original MCI. As can be seen, when compared to the circularity assessed using the original MCI method, the modified MCI method shows higher water and nitrogen circularity values for all four alternatives. This higher circularity is because of the consideration that both the water taken up by the crops and the water infiltrating underground contribute to the circular economy. Groundwater infiltration of the irrigation water may increase the cost for the farmers. However, it is known that improving the water use efficiency of irrigation often comes at the cost of groundwater recharge (Ebrahimi et al., 2016; Jiménez-Martínez et al., 2010; Xu et al., 2010). So, from the perspective of resource availability alone (despite

Table 3										
The circularity	of	the	alternatives	using	the	original	and	the	modified	MCI
methods										

Alternatives	Original MCI	results	Modified MCI results		
	Water circularity (%)	Nitrogen circularity (%)	Water circularity (%)	Nitrogen circularity (%)	
Freshwater irrigation + industrial fertilizer (FW 3)	0	0	3	41	
Fertigation every three days (TW_3)	41	29	44	73	
Fertigation every ten days (TW_10)	42	29	50	71	
Fertigation + drainage recycling (TW_10_DWR)	46	37	50	78	

the increased economic costs for the farmers), groundwater recharge does contribute to water circularity. A higher nitrogen circularity is because of the consideration that the plant uptake of nitrogen is restorative and  $N_2$  emission is a regenerative flow.

As seen in Table 3, water circularity improved from 3 % to 44 % because of switching to reused water. Nitrogen circularity improved from 41 % to 73 % when replacing industrial fertilizers with TW-N. A lower fertigation frequency improved water circularity from 44 % to 50 % while reducing nitrogen circularity from 73 % to 71 %. The water circularity improvement is due to lower evaporation losses, and the reduced nitrogen circularity is due to a lower crop nitrogen uptake. Lastly, collecting drainage water for reuse along with the runoff nitrogen improved nitrogen circularity from 71 % to 78 % because this prevented the dissipation of the TW-N. However, this intervention did not affect the water circularity because it was assumed that the uncollected drainage contributes to groundwater recharge.

The nitrogen and water circularities for the alternatives are shown in Fig. 5. Switching from FW and industrial nitrogen fertilizers to TW fertigation leads to the largest circularity improvement. Reducing fertigation frequency improves water circularity but reduces nitrogen



**Fig. 5.** Water and nitrogen (N) circularities for different fertigation schedules and drainage management using the modified MCI method. Whereas, freshwater irrigation every three days (FW\_3) has the lowest water and N circularities of 3 % and 41 %, treated water irrigation every ten days with drainage water recycling (TW\_10\_DWR) leads to the highest water and N circularities of 50 % and 78 %. When comparing treated water Irrigation every three days (TW\_3) with every ten days irrigation (TW\_10), it was found that the water circularity is improved from 44 % to 50 % with a lowered irrigation frequency, but the N circularity is reduced from 73 % to 71 %.

circularity. However, the decrease in nitrogen circularity can be offset if the nitrogen in the drainage water can be collected for reuse. The complete results can be found in the supplementary material.

# 4. Discussion

When comparing the circularity values of the original MCI with the new approach, the circularity of the fertigation case study was found to be lower using the original MCI method (e.g., 42 % water circularity for TW\_10 using the original MCI compared to 50 % using the modified MCI). The reason is that in the original method, only recycling/reuse of resources for human functions can be considered circular since the original MCI and most other methods were developed for the technical cycle resources. This is remedied by the modified approach to the MCI assessment.

The first factor leading to a higher water circularity of the modified MCI is the consideration of groundwater infiltration as a regenerative flow. Treated wastewater fertigation is mostly practised in arid and semi-arid regions of the world (Elgallal et al., 2016; Farhadkhani et al., 2018). Additionally, irrigation water is a major source of groundwater recharge flows, especially in arid and semi-arid regions (Jiménez-Martínez et al., 2009; Qin et al., 2011), contributing to future water availability. Thus, groundwater infiltration should be considered a contribution to circularity, even though it is an irrigation loss. Through the definitions provided in this paper, the groundwater infiltration flow falls under a regenerative type flow.

A disadvantage of the new method is that it does not account for the quality of the infiltration water. Reuse of treated wastewater for fertigation can lead to excess biogenic compounds and pharmaceuticals leaching into groundwater (Chojnacka et al., 2020). The impact of water quality should also be accounted for when considering the holistic sustainability impact of fertigation, but the method only deals with the

quantity of water since the focus is on the circularity aspect alone.

The second factor is crop water uptake. The total crop water uptake comprises the water in the edible as well as the non-edible parts. Here, it was assumed that the water flowing into the edible part is a restorative flow because this water will be directly used for human consumption. Further, it was assumed that the rest of the water would remain in the soil because the non-edible parts of the crop could be cut and left on the soil. Both of these are simplifications as some water will evaporate, and more accurate models are required to quantify such losses. For now, the total plant uptake of water is considered to be a restorative flow.

Similarly, the nitrogen circularity values from the modified MCI are higher compared to those from the original MCI (e.g., 29 % nitrogen circularity for TW\_3 using the original MCI as compared to 73 % using the modified MCI). The reason for this is the crop uptake of nitrogen. The nitrogen uptake to the edible part of the crop is a restorative flow because this nitrogen is meant for direct human use. A large quantity of nitrogen taken up by the non-edible part of a crop mostly remains in the soil after harvest (Fan et al., 2014; Gao et al., 2022; Poudel et al., 2001) and hence may be available for the subsequent crop (Kolberg et al., 1999; Poudel et al., 2001). Enhancing plant uptake of nitrogen is necessary for sustainability because the uptake can prevent nitrogen dissipation (Chen et al., 2019; Dimkpa et al., 2020).

It may be a simplification to assume that all of the nitrogen uptake remains in the soil after harvesting. Using more accurate nitrogen fate models is recommended to estimate exactly how much of the plant uptake nitrogen remains in the soil after harvest, but, for now, the plant uptake of nitrogen can be considered a restorative flow.

With the modifications to the MCI method, the circularity assessment was aligned better with sustainability in two ways. Firstly, by defining regeneration as the return of resources to the state in which the resources were appropriated from the natural environment, it was ensured that flows such as groundwater infiltration and  $N_2$  emissions count towards circularity improvement even though they are fertigation losses. Furthermore, crop uptake of water and nitrogen was considered to be restorative flows which means that maximising these flows also translates into improved circularity. This is logical because increasing the crop uptake of these resources can improve agricultural productivity and reduce losses.

Secondly, the circularity assessment was based on a resource flow model. Recycling a biogeochemical resource does not necessarily lead to a high circularity because of potential environmental losses such as the evaporation of reused water. A drastic reduction in river flows due to the growing consumptive use through evapotranspiration is a well-known phenomenon (Falkenmark and Lannerstad, 2005; Zisopoulou and Panagoulia, 2021). Without estimating environmental losses, which depend on local conditions, one risks an inaccurate assessment. It was shown how factors such as fertigation schedule and drainage water management could affect the circularity results.

Although the new approach was demonstrated on the short-timescale biogeochemical resources involved in treated wastewater fertigation, the approach can be applied to any resource recovery solution related to the water sector that deals with biogeochemical resources. This is because no restrictions were introduced by the modification presented in this study; rather, only the scope of application of the MCI method was extended by introducing some details related to the shortterm biogeochemical cycles.

Circularity values of fertigation are affected by the water and nitrogen fates which in turn are affected by the fertigation schedule and possibly other factors such as climate and rainfall. However, it was found that changing the fertigation frequency has opposite effects on water and nitrogen circularity values. The water circularity was found to be 50 % when the field was irrigated every ten days as compared to 44 %for every three days irrigation. Evaporative losses increase with a higher irrigation frequency (Mermoud et al., 2005; Mukherjee et al., 2010) since a smaller depth of water applied with a higher frequency leads to superficial wetting of soil, causing high evaporation (Mermoud et al., 2005) and lower infiltration, thus reducing circularity. Interestingly, the opposite effect of fertigation frequency is observed for nitrogen circularity which decreases from 73 % to 71 % when shifting from a three-day to a ten-day interval. This effect may be because crop uptake of nitrogen is known to increase with higher frequency fertigation of the tomato crop (Farneselli et al., 2015). The opposite effects on circularity based on the fertigation frequency means that careful planning is necessary to maximise both the circularities of water and nitrogen. This also means that in some cases, recovering nitrogen from wastewater and using it separately from irrigation water may be advisable for optimal circularity.

Collecting drainage water for reuse did not show any effect on the water circularity, which remained at 50 %. This is because water drained from the irrigated field was considered to be contributing to circularity regardless of being collected. In arid and semi-arid regions, where fertigation is most practised, infiltrating irrigation water is one of the major groundwater recharge flows (Ebrahimi et al., 2016; Jiménez-Martínez et al., 2010; Xu et al., 2010) and therefore, this flow was considered to be regenerative. If part or whole of the infiltrating water is collected for reuse using controlled drainage methods, then the collected flow will count as a restorative flow. In either case, the same water circularity is achieved. On the contrary, nitrogen circularity improved from 71 % to 78 % for the TW\_10\_DWR alternative. Collecting drainage water for reuse is known to reduce nitrogen loss (Reinhart et al., 2019; Williams et al., 2015) and thus contributes to improved nitrogen circularity, as confirmed by the assessment.

# 5. Conclusions

This study aimed to develop a new and improved circularity assessment approach for resource recovery solutions in the water sector. The novel approach resulted in higher circularity values compared to the original MCI method. This was because the new approach accounts for the fact that certain biogeochemical flows commonly classified as losses can contribute towards circularity. The application of this method to a real-life fertigation case study confirms that water circularity can be significantly improved with treated wastewater fertigation, especially with a low-frequency schedule. However, irrigation also leads to substantial water losses, mainly in the form of evapotranspiration and infiltration. While some of these losses (e.g., evapotranspiration) reduce water circularity, other losses (e.g., groundwater infiltration) may even contribute to water circularity. To ascertain the losses that are to be considered circular flows, the new definitions of restorative, regenerative, and linear flows provide a guideline. Further, using nitrogen contained in treated wastewater as a substitute for industrial fertilizers improves nitrogen circularity. However, contrary to water circularity, a lower application frequency can decrease nitrogen circularity by reducing its crop uptake. Nitrogen lost with infiltrating water should be collected for reuse, further improving nitrogen circularity. Summing up, a high-frequency application of TW-N along with drainage water collection can improve nitrogen circularity.

This method treats all irrigation water infiltrating underground as a desirable flow without accounting for its potentially low water quality. Future studies should explore how to quantify the effect of different infiltration water qualities on the sustainability of the groundwater and include this factor in the circularity assessment method. Further, the case study system boundary only included the irrigation field. Expansion of the system boundary to include the WWTP is recommended for future work. To conclude, the new approach to circularity assessment works well and can help to optimise the resource recovery solutions in the water sector. Although more accurate resource flow models and further discussion on restorative and regenerative flows are needed, the new approach is a crucial step towards ensuring a more circular and sustainable water sector.

# CRediT authorship contribution statement

Anurag Bhambhani: Conceptualization, Methodology, Formal analysis, Writing – original draft, Writing – review & editing. Zoran Kapelan: Conceptualization, Methodology, Writing – review & editing, Supervision, Project administration, Funding acquisition. Jan Peter van der Hoek: Conceptualization, Methodology, Writing – review & editing, Supervision, Project administration, Funding acquisition.

# 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.

# Data availability

Data used has been shared as supplementary material.

## Acknowledgements

This work is part of the project WIDER UPTAKE (www.wider-uptake. eu). This project has received funding from the European Union's Horizon 2020 - Research and Innovation Framework Programme under grant agreement No 869283. This article reflects only the author's view. The Commission is not responsible for any use that may be made of the information it contains.

# Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.166520.

### A. Bhambhani et al.

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#### A. Bhambhani et al.

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