



## Shifting the imbalance: Intentional reuse of Dutch sewage effluent in sub-surface irrigation



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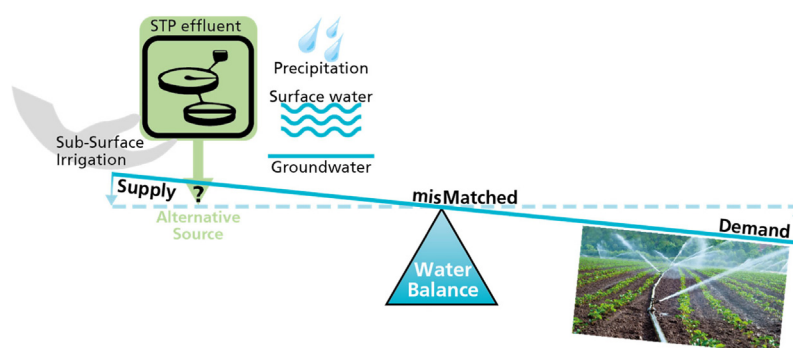
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### HIGHLIGHTS

- Possible alternative water resources are considered in order to meet the current and future water demand.
- Direct intentional reuse of STP effluent can satisfy a significant amount of the Dutch agricultural water demand via SSI.
- Prolonged SSI can elevate groundwater levels directly and indirectly via reduced groundwater abstraction.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Worldwide, agricultural irrigation currently accounts for 69% of freshwater withdrawal. Countries with a temperate climate, such as the Netherlands, experience periodic freshwater shortages in agriculture. The pressure on available freshwater will increase due to climate change and a growing demand for freshwater by e.g. industrial activities. Possible alternative water resources are considered in order to meet the current and future water demand. In this study we explore where, and how much, sewage treatment plant (STP) effluent can directly be reused in agricultural sub-surface irrigation (SSI) during an average and a dry season scenario, for all active (335) Dutch STPs. SSI systems may have a higher water demand as part of the STP effluent is transported with groundwater flow, although aboveground irrigation has a loss of water due to interception. Furthermore, such aboveground irrigation systems provide direct contact of crops with irrigation water. SSI systems provide a soil barrier which may function as a filter and buffer zone. In the Dutch situation, direct intentional reuse of STP effluent can fulfill up to 25% of croplands SSI water demand present within a five-kilometer transport buffer from the STPs during an average season and 17% during a dry season. Hereto, respectively, 78% and 84% of the total available Dutch STP effluent would be used. Thus, the intentional direct STP effluent reuse in agricultural SSI has the potential to satisfy a significant amount of the agricultural water demand at a national scale, presuming responsible reuse: safe applications for humans and environment and no limiting effects on water availability for other actors.

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## 1. Introduction

Globally, there is an increasing mismatch between the demand for and availability of freshwater resources (UN-Water, 2018). The main causes for water scarcity are interlinked, and include changes in water availability due to climate change, increases in water withdrawal for food production and for other economic activities such as industrial cooling water (FAO, 2016a). When water supply is scarce compared to demand, the question of how water is allocated becomes important (Bijl et al., 2018). Agricultural irrigation currently accounts for 69% of freshwater withdrawal worldwide (FAO, 2016b) which is not only an issue for regions with a high water stress index (WSI), but also low WSI regions with intense agriculture suffer from frequent non-potable freshwater shortages (Voulvoulis, 2018). At the same time the discharge of conventional sewage treatment plants (STPs) affects the receiving surface water quality (Johnson et al., 2020; Schwarzenbach et al., 2006), as these STPs are not optimized for the removal of poorly monitored and unregulated compounds, named contaminants of emerging concern (CoECs). Especially during low flow conditions, with usually high irrigation demand, surface water can consist primarily of effluent (Sousa et al., 2017; Yadav et al., 2017). Water from these streams is in many cases directly applied to crops by sprinkler and aboveground drip irrigation, resulting in the unintentional direct exposure of crops to pathogens and chemical micro-pollutants (Beard et al., 2019). Therefore, our research aims to analyze whether water scarcity in the agricultural sector and water pollution due to STPs effluent discharge can outbalance each other through the intentional direct reuse of STP effluent in controlled drainage systems.

Controlled drainage systems allow to retain groundwater within agricultural parcels; groundwater levels and soil moisture conditions can be actively regulated (Ayars et al., 2006). Introducing water turns a controlled drainage system into an infiltration system, which is called groundwater-fed irrigation or sub-surface irrigation (SSI). The goal of SSI systems is to raise the groundwater level and to improve soil moisture conditions for plant growth through capillary rise. Two major advantages that SSI with STP effluent via a controlled system may have compared to sprinkler irrigation, are (i) lower human health risks for workers due to no direct contact with the STP effluent, and (ii) optimal use of soil processes that stimulate degradation of CoECs (Narain-Ford et al., 2020). In addition, SSI is less-time-variable compared to aboveground irrigation and can sustain crops with high water requirements (Cucci et al., 2016; Machado and Serralheiro, 2017). Application will be limited to regions where SSI can raise the groundwater level such that the soil moisture availability in the root zone increases (Wada et al., 2014), so in regions with deep groundwater levels SSI will be no option. Moreover, there are uncertainties concerning the environmental and public health implications which are associated with reusing STP effluent for SSI in agriculture (Delli Compagni et al., 2020). Numerous laboratory, field and modelling studies describe the potential of soil-passage processes such as (bio)transformation to lower CoEC concentrations and sorption to reduce the mobility and concentrations in the (ground)water (Christou et al., 2019; Gonzales-Gustavson et al., 2019; Khalid et al., 2018; Blum et al., 2018). Accordingly, utilizing SSI as method of supply can improve water use efficiencies and may aid in the natural purification of these CoECs, although critical knowledge gaps remain (Narain-Ford et al., 2020).

So far, the direct intentional reuse of STP effluent has not reached its full potential in the European Union (EU) (Voulvoulis, 2018). The EU as the world's largest importer and exporter of agricultural products, with an estimated agricultural net value in 2017 of €432.6 billion, only intentionally reuses 2.4% (1,322 Mm<sup>3</sup>/year) of its STP effluent through mainly aboveground irrigation techniques (Eurostat, 2018; FAO, 2016b). Spain accounts for about one third of this (496 Mm<sup>3</sup>/year), followed by France (411 Mm<sup>3</sup>/year) (FAO, 2016b). Noteworthy, Cyprus reuses more than 70% of their produced STP effluent; however, this only accounts for 22 Mm<sup>3</sup>/year, less than 2% at a European level. In other EU member states,

STP effluent is reused on a smaller scale. Apart from Spain, France, Cyprus, Greece, Italy and Portugal there are no requirements among the EU member States on water reuse in national legislation or in non-regulatory standards (Joint Research Centre, 2017). Aquifer recharge (by surface spreading or direct injection) is only considered as a permitted use in Spain, Cyprus and Greece (Drewes et al., 2017). The EU recognizes the potential of STP effluent reuse in agriculture and recently the parliament approved the Water Reuse Regulation including safety requirements for the first time for STP effluent (European Commission, 2020). These new requirements of intentional use of STP effluent are expected to stimulate awareness around the prevailing unintentional reuse of STP effluent in agriculture and associated risks.

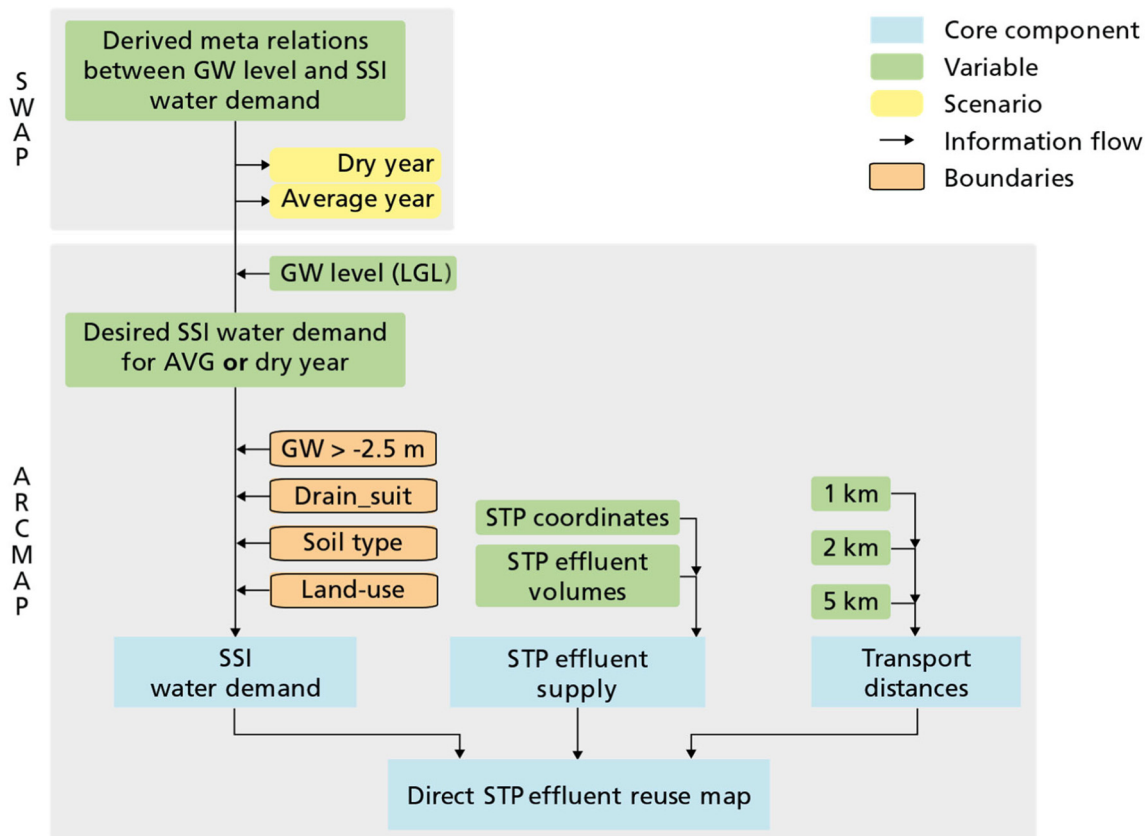
In order to answer the question how many croplands can be supplied with STP effluent through SSI, here the intentional direct STP effluent reuse in SSI to satisfy the water demands on regional and national scale was analyzed. The Netherlands, a densely populated country with 1.9 million hectares (Mha) cropland and well distributed STPs across the country, is selected as case study. As yearly average, the total sprinkler irrigation water demand in the Netherlands was estimated to be 144 million m<sup>3</sup> (CBS, 2016), with peaks during dry years up to 256 million m<sup>3</sup> (van der Meer, 2016). The yearly national annual STP effluent (1.9 billion m<sup>3</sup>/y) is much higher than the annual estimated water demand for aboveground irrigation systems. Compared to aboveground irrigation techniques, SSI requires a lot of water for raising groundwater to a desired level, while less irrigation water is lost to the atmosphere through evaporation of interception water (Narain-Ford et al., 2020). However, since most irrigation is needed in the summer months in specific regions with sandy soils (Witte et al., 2019), local and temporal availability might not be able to meet the demand. In addition, aboveground irrigation techniques result in direct exposure of crops to STP effluent. In this context, an exploratory spatial analysis considering local STP effluent volumes related to local SSI water demands during a dry and an average season was performed.

## 2. Materials and methods

A Dutch STP effluent reuse map representing where, and how much, STP effluent can directly be reused in agricultural SSI was created in ArcGIS10.5. at a 25m x 25m resolution. The site-independent workflow for the creation of this direct STP effluent reuse map is presented in Fig. 1. Each component of this workflow is discussed in the following paragraphs. Briefly, this map combines croplands and local SSI water demand estimations with the available local STP effluent volumes. In addition, it includes several hydrological conditions, such as groundwater levels and soil physical properties to determine the suitability of SSI as method of supply. Finally, three different water transport distances are incorporated to simulate the potential of effluent reuse.

### 2.1. Sub-surface irrigation water demand

In order to acquire SSI water demand on a national scale, we adapted the tool of Bartholomeus and Witte (2013) to derive soil-specific transfer functions (hereafter meta-relations) between groundwater level characteristics (mean Lowest Groundwater level: LGL) and SSI water demand in the Soil Water Atmosphere Plant model (SWAP, Kroes et al., 2017). This tool considers the detailed processes in the soil-water-plant-atmosphere system that are relevant for SSI and translates process-based simulations to meta-relations. These meta-relations can be easily applied to estimate SSI water demand from the output of regional and/or national hydrological models. Within SWAP three main Dutch soil types, i.e. peat, sand and clay, were collected from the Soil Physical Unit map, that can further be divided into 21 sub-soil types (Wösten et al., 2013). For each of the 21 sub-soil types, a linear meta-relation was estimated between (i) mean Lowest Groundwater level (LGL) in the situation without SSI and (ii) the amount of water needed to reach the groundwater level where crops can extract water through capillary rise,



**Fig. 1.** Workflow for the creation of a direct STP effluent reuse through SSI map. Core components are the building blocks for the creation of this map, variables are prone to change over time, scenarios refer to the weather conditions, boundaries implicate the suitability of Dutch croplands for SSI.

i.e. SSI water demand. These meta-relations were derived for 30-year average conditions and for the dry year 2003 (Fig. 1). We focused on the time of the year with an irrigation water demand, i.e. the growing season from 1st April until 1st October.

Fifteen SSI water demand simulations were run for the selected 21 sub-soil types in SWAP for the current climatic conditions, i.e. a period of 30 years (1981–2010) using: i) daily meteorological data from the Royal Meteorological Institute (KNMI) of De Bilt in the centre of the Netherlands ii) solely grass as crop and iii) for a range of hydrological boundary conditions (Table 1).

Once meta-relations have been derived for the soil types and climate in a specific area, modelled groundwater levels for large grids can be

transferred to SSI water demand for each grid cell. Spatial data on groundwater levels obtained from the National Water Model (NWM) Instrument, with a spatial resolution of 25m x 25m (Bos-Burginger et al., 2018), and only for cells with agriculture, were combined with the derived meta-relations to acquire a spatial map of SSI water demand. The vector-based file containing the geographical position of Dutch croplands, i.e. grass, maize, potatoes, sugar beets, grains, bulbs and others, in 2018 was extracted from the National Land-use 6 (LGN6) database with a resolution of 250m x 250m. In ARCMAP10.5 this resolution was converted to 25m x 25m.

2.2. STP effluent volumes

The available Dutch STP effluent discharge volume data, from 2010 until 2016, were retrieved from the Centre for Big Data Statistics (CBS) database and coupled with the latitude and longitude coordinates that were retrieved from the EU dissemination platform related to the Urban Waste Water Treatment Directive (<http://uwatd.oieau.fr/>). Within these years no extreme dry or wet years were observed (KNMI, 2018). All active Dutch STPs, in total 335, were compiled. For each STP, we assumed that effluent volumes were equally distributed throughout the year.

2.3. Transport distances

Three random maximum radial transport distances from STPs to croplands were selected; i.e. one, two and five kilometers. Longer distances contributed to many overlapping areas, and might lead to unfeasible transportation cost (Dermody et al., 2018). In doing so, only direct reuse of STP effluent was considered and indirect reuse (de facto reuse) from surface water further downstream was excluded from the analysis. Furthermore, the SSI water demand calculated per grid cell

**Table 1**

Ranges of input values for SWAP in order to generate a range of hydrological boundary conditions for which the SSI water demand is simulated. For used SWAP input files and more details on the used SWAP parameters we refer to the electronic appendix A.

SWAP parameter	Description	Value/Range
<i>Parameters to simulate bottom boundary conditions</i>		
RIMLAY	Vertical resistance of aquitard	10–2,500 d
SHAPE	Shape factor to derive average groundwater level	0.1–0.9 [–]
AQAVE	Average hydraulic head in underlying aquifer	125–250 cm--soil surface
AQAMP	Amplitude hydraulic head sinus wave	0.1–75 cm
AQTMX	First time of the year with maximum hydraulic head	60–120 [d]
QBOT4	Extra groundwater flux	–0.1–0.1 cm/d
<i>Parametrization of surface water system</i>		
L1	Distance between surface water units	50–500 m
Factor_L	Factor to calculate drainage resistance from L1 (Van Der Gaast et al., 2006)	1–3
ZBOTDR1	Bottom depth of surface water units	120–250 cm--soil surface

**Table 2**  
Croplands captured within the one, two- and five-kilometer buffer transport distance.

Buffer [km]	Croplands within the buffer <sup>a</sup>	
	Number of croplands	Surface area (km <sup>2</sup> )
1	16,054 (3%)	372 (2%)
2	67,027 (11%)	1580 (10%)
5	341,181 (55%)	8655 (52%)

<sup>a</sup> 100% are 620,379 croplands with SSI demand and a surface area of 16,508 km<sup>2</sup>.

was averaged for each individual plot of cropland, in order to avoid: i) multiple SSI water demands within one cropland and ii) only supplying parts of a cropland with irrigation water when it is located on the border of the selected transport buffer.

The ARCMAP10.5 near\_rank analysis was used to calculate the number of croplands, including their surface area, within each transport buffer. This analysis allows for the ranking of the distance between a cropland and one or more STPs based on their proximity within the one-, two- or five- kilometer transport buffer.

On a national scale the Netherlands consist out of 620,379 croplands, which corresponds to 16,508km<sup>2</sup> agricultural land-use (the electronic appendix B). The one- and two-kilometer transport buffer cover 2% and 10%, respectively, of the total cropland surface area. In these two buffers every cropland falls into only one STP transport buffer. The five-kilometer buffer can capture 55% of Dutch croplands which equals 52% of Dutch cropland surface area (Table 2). For the five-kilometer transport buffer croplands fall into a maximum of seven STPs transport buffer. Here, the STP closest to a cropland was appointed to rank\_1. The second closest STP of a cropland was assigned rank\_2, and so on. If the STP closest to a cropland (rank\_1) could not fulfill the SSI water demand, the second closest STP (rank\_2) was used. The third until the seventh closest STP were not used to fulfill the SSI water demand, due to their insignificant contribution (Table 3). The STP effluents are equally distributed across the water demand by all croplands in the supply area.

### 3. Results

This section presents density maps for: 1) the estimated Dutch SSI water demand per cropland, 2) the distribution of Dutch STP effluent and 3) the amount of Dutch SSI water demand fulfilled by the available Dutch STP effluent within a one, two- and five-kilometer transport buffer.

#### 3.1. SSI water demand

The 30-year average SSI water demand for all croplands is 7.05 billion m<sup>3</sup>/yr. The dry year 2003 had an SSI estimated water demand of 10.50 billion m<sup>3</sup>/yr. The one- and two-kilometer transport distances capture less than 10% of the SSI water demand. A transport distance of five-kilometer from an STP captures around 50% of this national SSI water demand in both a dry and an average year (Table 4).

The east of the Netherlands, adjacent to Germany, is characterized by sandy soils (Wösten et al., 2013) with low groundwater levels, that directly correspond to the highest Dutch SSI water demands. The SSI water demand distribution is presented in Fig. 2.

**Table 3**  
Croplands captured by one or more STPs for the five-kilometer buffer.

Rank	Number of croplands	Surface area (km <sup>2</sup> )
1	341,181	8655.15
2	84,086	1840.14
3	22,281	421.05
4	5491	87.98
5	1525	28.72
6	247	5.74
7	16	0.65

**Table 4**  
SSI water demand captured within the one, two and five kilometer transport buffer.

Buffer [km]	Average year SSI demand (m <sup>3</sup> ) <sup>a</sup>	Dry year SSI demand (m <sup>3</sup> ) <sup>a</sup>
1	151,366,991	229,806,460
2	642,421,126	975,055,481
5	3,565,379,507	5,381,250,253

<sup>a</sup> 100% is 7,049,861,265 m<sup>3</sup> for an average year and 10,503,394,180m<sup>3</sup> for a dry year.

#### 3.2. STP effluent water supply

The 335 active Dutch STPs treat a wastewater volume of 1.9 billion m<sup>3</sup>/y. This equals 0.95 billion m<sup>3</sup> per growing season (6 months) assuming STP effluent is constant over the year. The distribution of the total available Dutch STP effluent considering croplands as land-use is presented in Fig. 3. The electronic appendix B presents the distribution of Dutch croplands and the STP effluent distribution independent of croplands.

#### 3.3. Fulfilled water demand

Within a five-kilometer transport buffer distance of STPs, 25% of the national cropland water shortage can be reduced via SSI during an average growing season and 17% during a dry season. These percentages correspond to 78% and 84%, respectively, of the national STP effluent volume being reused. A transport buffer distance less than 5 km can satisfy fewer croplands (Table 5). Regarding the column 'fulfilled water demand' of Table 5 it must be remarked that croplands not within the selected buffer distances of the 335 active Dutch STPs remain unfed. Also, noteworthy, only 5.61% of STP effluent from the six-month average (Fig. 3) is beyond the five-kilometer STP buffer transport distance and thus cannot be used to fulfill cropland demands. Regarding the column 'remaining STP effluent' it must be emphasized that STP effluent currently already has a function: it feeds local surface waters. By reusing effluent for SSI, the direct discharge to surface waters will decrease, however the baseflow may increase due to shallower groundwater levels. All in all, for each specific case it is needed to determine the volume of effluent that can be used responsibly, without causing negative effects to other functions.

Fig. 4 displays the SSI water demand fulfilled by STPs for a five-kilometer spatial distance in a dry and an average season. In a dry year scenario, most croplands with sandy soils (in the east of the Netherlands) can only be supplied up to 10% (0.10 in fractions) of their SSI water demand. Allocating these STP effluents to neighboring croplands that can be supplied by more than 10%, 30% and 50% may be an interesting development.

Communities in areas with a high SSI water demand produce relatively small amounts of STP effluent; whereas the urbanized areas with a low cropland water demand produce larger amounts of STP effluent. This is especially the case for the middle-west of the Netherlands, where a five-kilometer buffer distance can fully capture the SSI water demand during an average season. Moreover, a surplus of more than 10 times the STP effluent reused for SSI remains. In these areas longer transport distances could be considered.

The one- and two-kilometer transport buffer distances cover a small portion (less than 10%) of croplands surface area. For their SSI water demand fulfilled by STP effluent density maps we refer to the electronic appendix C.

## 4. Discussion

#### 4.1. Uncertainties and limitations of approach

Our results indicate that 341,181 croplands with a surface area of 8,655 km<sup>2</sup> can be captured by a five kilometer transport buffer distance. This equals 55% and 52%, respectively, of the national amount of croplands (620,379) and the national cropland surface area (16,508km<sup>2</sup>). The STP



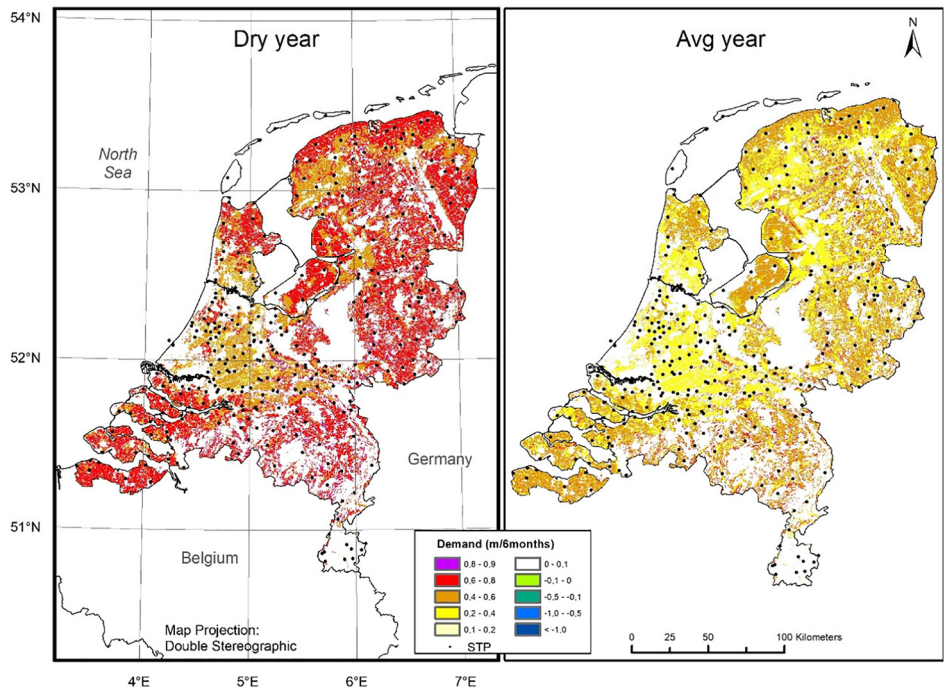


Fig. 2. SSI water demand [m] per 25m grid cell for SSI suitable croplands.

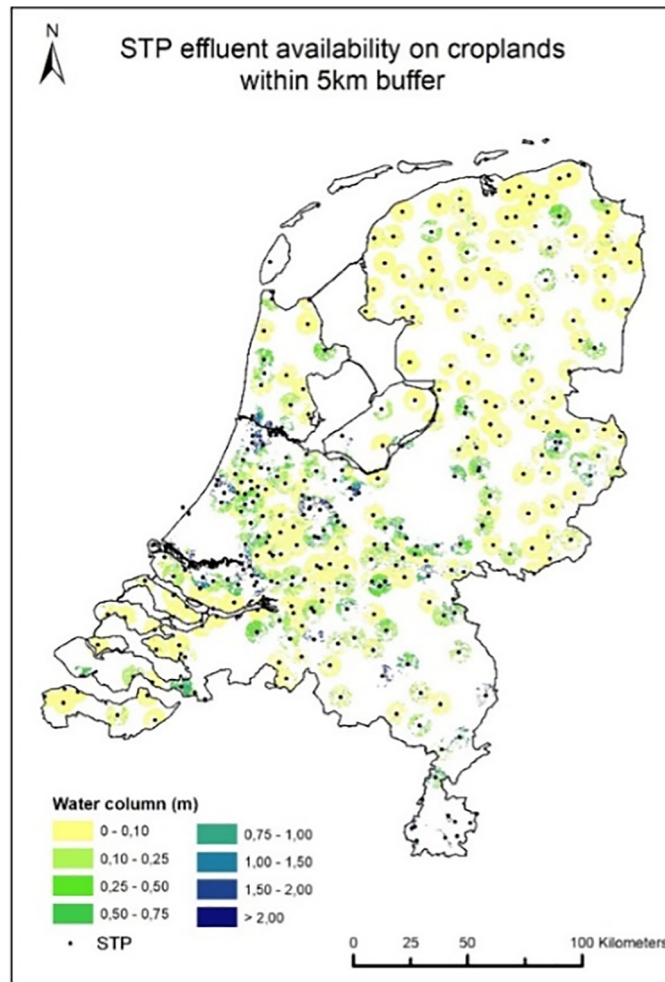


Fig. 3. Distribution of Dutch STP effluent volumes per growing season.

**Table 5**  
Percentage of croplands SSI water demand satisfied within three buffer distances and corresponding STP effluent reused.

Buffer [km]	Fulfilled water demand within buffer		Remaining STP effluent after SSI <sup>a</sup>	
	Average	Dry	Average	Dry
1	100%	100%	84%	76%
2	100%	81%	32%	17%
5	25%	17%	22%	16%

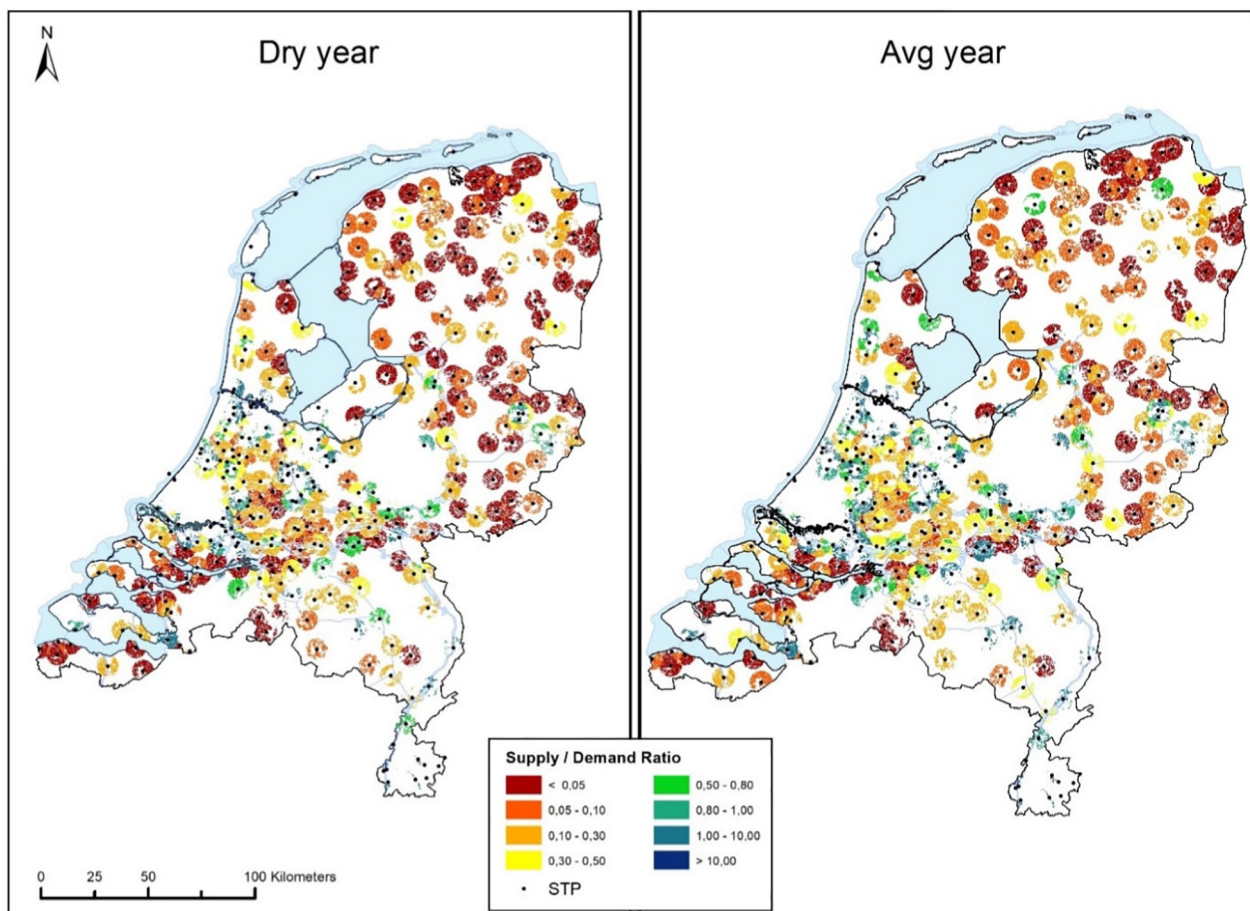
<sup>a</sup> 100% is the 6 months total of Dutch STP effluent of 0.95 billion m<sup>3</sup>/y.

effluent volume (0.95 billion m<sup>3</sup>) available during the six month growing season can supply a significant part of the Dutch croplands SSI water demand (25% during an average season and 17% during a dry season) by the intentional direct STP effluent reuse through SSI. A transport distance of 10 km was considered feasible by Orange County Water District (OCWD, 2019). Therefore, a surplus of STP effluent (see Fig. 4) may be utilized towards croplands outside of the five kilometer transport buffer zone based on the assumptions made. Several uncertainties surrounding these assumptions can be mentioned e.g.; these analyses provide the average for the national SSI water demand in a dry and average year. Local water requirements for agriculture and other sectors may have significant variations and depend upon a more detailed analysis. In addition, neither groundwater protection zones nor irrigation water quality requirements were considered in the current analyses, while SSI with STP effluent might bear the risk of groundwater and crop contamination (Barbagli et al., 2019). Furthermore, required groundwater recharge to satisfy the SSI water demand was solely estimated for grass yield. Irrigation water requirements differ per crop type and therefore,

simulated water demands may be over- or underestimated. Finally, we assumed that 100% of STP effluent is available for SSI. As minimum stream flow conditions are required for ecosystem functioning in some basins not all STP effluent can be reused for irrigational purposes (Beard et al., 2019; Poff, 2018). All in all, a detailed analysis on water demand, water supply and water quality is required to assure responsible reuse for implementation in specific cases.

#### 4.2. Practical implications

Different disciplines are needed to explore the full extent of STP effluent reuse for the practice of responsible water reuse (Dingemans et al., 2020). Wastewater reuse, presuming safe application, can have a significant contribution to the agricultural water demand and may limit the pressure on freshwater resources. STP effluent is commonly indirectly and unintentionally reused in agriculture by irrigating with surface water in which STP effluent was discharged (Beard et al., 2019). Conventional STPs are not optimized for the removal of CoECs and their discharge will affect the receiving surface water quality (van Wezel et al., 2018). Especially during low flow conditions with usually high irrigation demand, surface water can consist primarily out of effluent. In these cases, having SSI as method of supply eliminates direct contact of crops and fieldworkers to STP effluent. Particularly in developing countries using this method of supply may better reflect the health benefits of no direct contact (Awad et al., 2019). Utilizing SSI instead of aboveground irrigation techniques may also aid in keeping the groundwater prolonged at a desired level and prevent salt water intrusion (Hack-Ten Broeke et al., 2016). In this analysis, we optimized SSI systems with transport lengths of maximum 5 km. In more arid areas water is already transported over longer distances through



**Fig. 4.** Fraction of the SSI water demand fulfilled by STP for a five-kilometer spatial distance in a dry and an average season.

aqueducts (i.e. canals, pipes). Hanasaki et al. (2018) compiled several of these major systems longer than 50 km in six different countries. This means that those systems are potentially feasible to transfer water in general, making SSI potentially interesting for larger areas.

## 5. Conclusion

SSI with STP effluent can supply a significant part of the agricultural water demand, while also maintaining desired groundwater levels. According to previous studies (Stuyt, 2013; Terink et al., 2013) the cost and implementation of such systems ranges between €200 and €270/ha per year.

As yearly average, the total sprinkler irrigation water demand in the Netherlands was estimated to be 144 million m<sup>3</sup> (CBS, 2016), with peaks during dry years up to 256 million m<sup>3</sup> (van der Meer, 2016). Therefore, sprinkler irrigation may be able to supply all Dutch croplands with STP effluent. However, such aboveground irrigation systems provide direct contact of crops and fieldworkers to STP effluent and they are not exempt from evaporation (Gunarathna et al., 2017). Indeed, SSI systems may have a higher water demand than aboveground irrigation systems, in this study the difference was approximately 50-fold. However, soils suitable for SSI (Narain-Ford et al., 2020) provide a saturated soil barrier which may function as a filter and buffer zone. Additionally, the infiltrated water that is not used by the plants recharges the groundwater, and so the water is not lost. Moreover, the buffer function of the subsurface may allow for a temporal storage of STP effluent during the winter season, presuming soils are suitable for SSI. It can also be expected that, based on the size of the area infiltrated with STP effluent during SSI, over a prolonged period the SSI water demand will diminish because of elevated groundwater levels. Though, here the balance between desired groundwater level and soil recovery through rainfed dilution should be guarded.

Our study may be characteristic for other parts of the world that have high population densities, suitable wastewater collection and treatment near agricultural areas, and suitable soil conditions. Improving the quality of the STP effluent by upgrading conventional STPs to include a tertiary treatment such as wetlands may also aid in lowering the health and environmental risks of STP effluent reuse (Nguyen et al., 2020). Withal, the extent to which SSI systems diminish and/or retain CoEC in croplands including an adequate risk assessment of SSI with STP effluent should be assessed in future studies in order to get a sound understanding of the possible opportunities and limitations of STP effluent reuse in such systems.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2020.142214>.

## CRediT authorship contribution statement

R.B., D.N., B.R., I.v.Z., T.t.L., A.v.W. and S.D. designed the study. R.B. performed the hydrological simulations. B.R., D.N. and I.v.Z. performed ArcGIS analysis. D.N. prepared the original draft of this paper. S.D., A.v.W., R.B., I.v.Z., T.t.L. and B.R. provided critical feedback to the original draft of this paper. All authors approve the final version of the paper.

## Declaration of competing interest

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

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