Recommendations to derive quality standards for chemical pollutants in reclaimed water intended for reuse in agricultural irrigation

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Keywords: Reclaimed water; Agricultural irrigation; Compounds of Emerging Concern; Quality Standard; Prioritisation; Treated wastewater reuse

Abstract The reuse of treated municipal wastewater (herein referred to as reclaimed water) in agricultural irrigation (RWAI) as a means to alleviate water scarcity is gaining increasing policy attention, particularly in areas where water demand mitigation measures have proved insufficient. However, reclaimed water reuse in practice is lagging behind policy ambition, with <2.5% of it reused in a European context. A key barrier identified as limiting its full valorisation is concern over its impact on human and environmental health. To address this concern, and to meet further objectives including achieving parity between current reclaimed water reuse guidelines operational in various Member States, the European Commission has proposed a regulation which identifies minimum quality requirements (MQR) for a range of microbiological and physico-chemical parameters but the inclusion of compounds of emerging concern (CECs) in terms of the determination of quality standards (QS) is missing. This paper reviews the existing pertinent EU legislation in terms of identifying the need for CEC QS for RWAI, considering the scope and remit of on-going pan-European chemicals prioritisation schemes. It also evaluates opportunities to link in with the existing EQS derivation methodology under the EU WFD to address all protection targets in the environmental compartments exposed via potential pathways of RWAI. Finally, it identifies the main data gaps and research needs for terrestrial ecosystems, the removal efficiency of CECs by WWTPs and transformation products generated during the wastewater reuse cycle.

1. Introduction

Over 2 billion people live in countries experiencing high water stress, with approximately 4 billion people experiencing severe water scarcity during at least one month of the year (WWAP, 2019). Whilst freshwater is relatively abundant in the European Union (EU), around 30% of the total European population, experienced water scarcity conditions in the summer of 2015 compared to 20% in 2014 (EEA, 2019). Whilst total water abstractions within Europe have decreased by an estimated 19% since 1990 (driven by efficiency gains likely to continue in the coming years), water stress hotspots are predicted to remain and even grow given continued pressures such as expanding urbanisation, increasing population and climate change (EEA, 2019). Forecasts such as these highlight the urgent need to utilise alternative water resources such as treated wastewater (referred to henceforth as reclaimed water reuse) when water efficiency, demand management and improved agricultural practices are not sufficient to prevent water scarcity.

A study by WWAP (2017) distinguishes three main types of reclaimed water reuse: 1) direct potable reuse; 2) indirect potable reuse; and 3) reuse for non-drinking purposes including agricultural irrigation. This study reports that the reclaimed water reuse in agriculture is an area of great potential and indeed, the European Environment Agency (EEA) report on European waters indicates that in the Spring of 2014, the agricultural sector used 66 % of the total water used in Europe (EEA, 2018). Whilst current EU legislation encourages reclaimed water reuse through the Urban Waste Water Treatment Directive (UWWTD, EEC 1991) and the Water Framework Directive (WFD, EU 2000), these legislative pieces only refer to reuse practices in brief without specifying conditions for reclaimed water quality for reuse practices. During the 2012 fitness check on EU freshwater policy (EC, 2012), industry stakeholders raised concerns about the lack of EU quality standards for reclaimed water reused in agricultural irrigation (RWAI), with potential impacts on the free movement of agricultural produce in the single market highlighted. Six Member States (MSs) (Cyprus, France, Italy, Greece, Spain and Portugal) have requirements in place which set-out quality requirements for reclaimed water reuse either in legislation or as non-regulatory standards, but these requirements vary significantly in terms of both parameters included and their associated values. Additionally, it should be noted that EU citizens in many MSs have expressed their concerns about water safety e.g. the European Citizens' Initiative "Right2Water", ECI R2W, 2012). and many international water reuse initiatives like in the US or Singapore have faced public opposition (Voulvoulis, 2018).

In response to identified concerns over variations in quality standards and associated implications for the transnational shipment of irrigated crops, the European Circular Economy Action Plan (EC, 2015) announced activities to facilitate reclaimed water reuse, including a proposal to develop legislation on minimum quality requirements (MQRs) for RWAI and groundwater recharge. In May 2018, the European Commission (EC) put forward a legislative proposal for a regulation of the European Parliament and of the Council on MQRs for reclaimed water reuse seeking to incentivise reuse, while ensuring a high level of protection of health and the environment (EU, 2018). Reclaimed water, defined as urban wastewater that has undergone treatment in a reclamation facility, will be used to irrigate food crops, processed food crops and non-food crops. The EC estimates that the proposal could enable the reuse of more than half of the current volume of water coming from EU wastewater treatment plants within irrigation, resulting in a reduction of water stress of >5%. Despite requesting over 400 amendments to the proposed text, the European Parliament adopted its first reading position on 12 February 2019 and in the Council, the proposal is being examined by the Working Party on the Environment. The main political issues that have emerged from the discussion include the degree of flexibility the EU instrument should offer to MSs and the stringency of the MQRs for reclaimed water quality (EPRS, 2018). The regulation proposal defines MQRs for microbiological (e.g. *Legionella, E. coli*) and physicochemical (BOD, TSS and turbidity) parameters. It states that competent authorities would have the possibility to impose additional requirements, based on a risk management plan submitted by the reclamation plant operator, or on the need to mitigate unacceptable risks to health or the environment. The legal proposal is based on a review of current knowledge pertaining to reclaimed water reuse developed by EC Joint Research Centre

(JRC, 2017). The development of the proposal adopted a tiered approach, requesting reviews of proposed MQRs from the EU Scientific Committee on Health, Environment and Emerging Risks (SCHEER) and the European Food Safety Authority (EFSA). Both (EFSA, 2017 and SCHEER, 2017) were of the opinion that the proposal provided insufficient protection to environmental and human health especially with regard to Compounds of Emerging Concern (CECs) and disinfection by-products associated with wastewater treatment plants (WWTPs). SCHEER recommended developing common criteria and detailed guidance on MQRs for priority CECs and EFSA identified the need to assess their impact on human, animal and environmental health. In addition, the JRC presented their findings and recommendations at several public events and scientific meetings. On various occasions, members of the COST Action ES1403 NEREUS also presented the current stage of knowledge concerning CECs and wastewater reuse in such meetings. The COST Action ES1403 established a multi-disciplinary network to determine which of the current challenges related to wastewater reuse are the most concerning in relation to public health and environmental protection, and how these could be overcome. A core activity of the network was the collaborative development of a framework to support the qualitative assessment of risks associated with reclaimed water reuse. At the final conference of the NEREUS COST Action in Limassol, Cyprus, in October 2018, a panel was organised to discuss the 'big unknowns concerning safe and sustainable wastewater reuse' and, more specifically, how effective the proposed MQRs are. The panel concluded that, in their current form, the proposed MQRs provided insufficient protection both to environmental and human health. This international discussion panel has been the starting point for a more in-depth consideration on how to address this identified knowledge gap on chemical pollutants, leading to the development of practical recommendations on how to prioritise substances and develop MQRs to support the safe use of RWAI.

2. Identification and prioritisation of chemical contaminants of concern

Several studies in the literature report the presence of a range of organic and inorganic contaminants in the effluents of urban WWTPs in the EU. For example, Aguayo et al (2004) identified more than 49 compounds in the organic fraction of effluents from seven urban WWTPs in close proximity to urban and industrial areas within the community of Madrid (Spain). Karvelas et al. (2003) investigated the occurrence and the fate of heavy metals (Cd, Pb, Mn, Cu, Zn, Fe and Ni) in the urban WWTP of the city of Thessaloniki (northern Greece) and found that 47–63% of influent concentrations of Cd, Cr, Pb, Fe, Ni and Zn remain in the treated effluent. According to Rizzo et al. (2019) urban WWTPs are not designed to remove CECs and secondary (e.g. conventional activated sludge process, CAS) and tertiary (such as filtration and disinfection) treatment processes are not effective in the removal of most of the CECs entering urban WWTPs. In order to identify which are the relevant substances that could be present in reclaimed water, we propose to identify the different sources of wastewater in the water reuse system as recommended in the Water Reuse Risk Management Plan defined in the Annex II of the EC reclaimed water reuse regulation proposal (EU, 2018).

2.1 Identification of candidate substances reaching urban WWTPs

2.1.1 Sources of wastewater and European policy landscape

The EC proposal (EU, 2018) focuses on treated wastewater as covered by the UWWTD (EEC, 1991) where urban wastewater is defined as domestic wastewater or the mixture of domestic wastewater with industrial wastewater and/or run-off rainwater (see Fig. 1). Domestic wastewater refers to effluents from residential settlements and services which originate predominantly from the human metabolism and from household activities. All substances contained in consumer products but also professionals’ products related to consumer services could be released into such wastewater. Industrial wastewater refers to discharges from premises used to carry out a trade or industry as described in Annex I of the Industrial Emissions Directive (IED; EU, 2010). All substances manufactured, formulated or used by the referenced categories of activities could be released in such wastewater. The UWWTD states that the discharge of industrial wastewater to urban WWTPs is subject to prior authorizations by MSs and sets the requirement to both protect the health of staff and ensure that discharges from these plants do not adversely affect the environment. However, the effectiveness of this Directive almost 25 years after its adoption is now questioned and its evaluation was recently initiated by the EC, to consider in particular to which extent its quality standards (both in relation to pollutants listed and limit values identified), reflect technological developments and meet today's challenges. Industry's wastewater releases into water bodies (direct releases) as well as wastewater releases into public sewage which ends within urban WWTP (indirect releases) are among the key aspects regulated by the IED (EU, 2010). All permit conditions must be based on the environmental protection level provided by the use of Best Available Techniques (BAT) with associated emission limit values identified for each installation. However, according to the BAT for common wastewater and waste gas treatment/management systems in the chemical sector (BAT, 2016), the main risk to be addressed when discharging industrial effluents to an urban WWTP is to ensure that pollution levels in the effluent will not damage or diminish sewer system performance. In order to reduce emissions to water, BAT involves the pre-treatment of wastewater that contains pollutants that cannot be dealt with adequately during final wastewater treatment. Associated emission levels to water (BAT-AELs) are recommended for indirect releases however, they are set for groups of chemicals (i.e. some heavy metals and adsorbable organically bound halogens) and not for individual substances making difficult to assess their efficacy to control the risk of prioritised substances identified for RWAI. Finally, the European Pollutant Release and Transfer Register Regulation (E-PRTR; EU, 2006) places a legal obligation on the EC and the MSs to establish a coherent, EU-wide pollutant register concerning emissions from industrial activities, including WWTPs. The E-PRTR is the largest industrial emissions database in Europe, containing data on more than 90 substances from 45 economic sectors. However, it is recognised that the current scope of the E-PRTR does not capture all pertinent industrial emissions to water as the substances covered have not been revised since the regulation was adopted and reporting is required only for certain activities, emission thresholds and urban WWTPs serving greater than 100 000 population equivalents (EEA, 2019). Stormwater run-off is not referred to in the UWWTD but, depending on land use and weather conditions, can be an important additional source of pollutants entering urban WWTPs (e.g. traffic-related activities, combustion products) as described by Lundy et al. (2011), Christoffels and al. (2016) and Brudler et al. (2019).

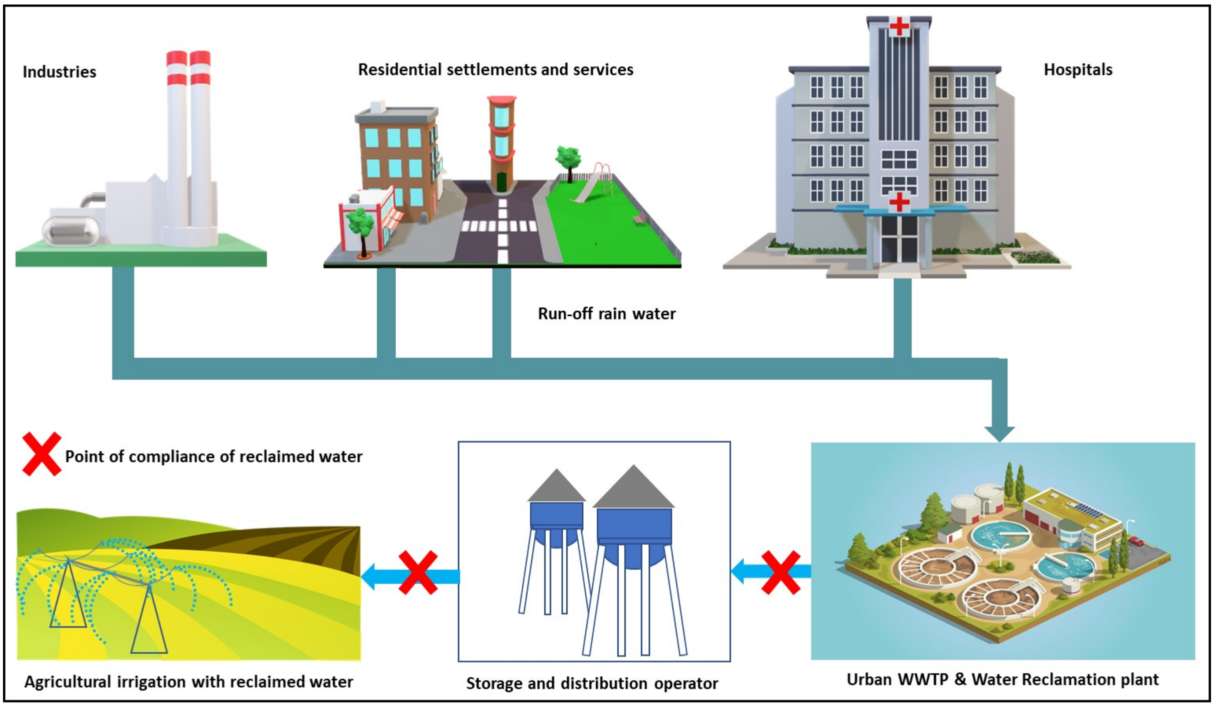

Figure 1. Sources of wastewater to urban WWTP and reclaimed water cycle for reuse in agricultural irrigation.

The management of industrial and urban wastewaters is also regulated indirectly by the WFD and the Environmental Quality Standards (EQSs) Directive (EU, 2008) amended by the Priority Hazardous Substances Directive (EU, 2013) which aim to ensure that all aquatic ecosystems achieve 'good chemical and ecological status'. However, the good chemical status of water bodies is defined in these Directives as compliance with quality standards established at EU level for only 45 Priority Substances (PSs) and certain other pollutants (including ubiquitous PBTs: persistent, bioaccumulative and toxic chemicals) and hence does not comprehensively address all pertinent CECs. In addition, the WFD establishes the principles to be applied by the MSs to develop EQSs for specific pollutants that are 'discharged in significant quantities' as forming part of the assessment of ecological status and hence provides an opportunity to address pertinent CECs at national level.

2.1.2 Potential chemical categories

Based on the identified sources of wastewater in urban WWTPs, the chemical categories that could potentially be present in wastewater streams arriving at a treatment and reclamation plant, the corresponding chemical legislation at an EU level and the European agencies responsible for their implementation have been identified (see Table 1). It must be noted that disinfection agents used at WWTPs and releasing by-products of growing concern are biocidal products regulated by the Biocidal Products Regulation (BPR).

Table 1: Chemical categories of compounds that can reach urban WWTPs with corresponding EU legislation and implementing authorities.

Chemical category	**EU legislation**	**EU implementing authorities**

Industrial chemicals	[a] REACH Regulation [b] CLP Regulation	European Chemicals Agency (ECHA)
Biocidal Products also called Biocides	[c] BPR Regulation [b] CLP Regulation	European Chemicals Agency (ECHA)
Human medicinal products also called Pharmaceuticals	[d] Medicinal products for human use Directive	European Medicines Agency (EMA)
Veterinary medicinal products	[e] Veterinary medicinal products Directive	European Medicines Agency (EMA)
Plant Protection Products (PPPs) also called Pesticides	[f] PPP Regulation [b] CLP Regulation	European Food Safety Authority (EFSA) European Chemicals Agency (ECHA)
Cosmetic products	[g] Cosmetics products Regulation	European Chemicals Agency (ECHA)
Food and Feed additives	[h] Food additives Regulation [i] Feed additives Regulation	European Food Safety Authority (EFSA)
Other substances like combustion products unintentional by-products of industrial processes (e.g. dioxins and furans)	No global legislation but some are covered by the [j]POPs Regulation	European Chemicals Agency (ECHA) for POPs

[a] Regulation (EC) No 1907/2006 of the European Parliament and of the Council of 18 December 2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals.

[b] Regulation (EC) No 1272/2008 of the European Parliament and of the Council of 16 December 2008 on Classification, Labelling and Packaging of substances and mixtures.

[c] Regulation (EU) No 528/2012 of the European Parliament and of the Council concerning the making available on the market and use

of Biocidal Products.

[d] Directive 2001/83/EC of the European Parliament and of the Council of 6 November 2001 on the Community code relating to medicinal products for human use.

[e] Directive 2001/82/EC of the European Parliament and of the Council of 6 November 2001 on the Community code relating to veterinary medicinal products.

[f] Regulation (EC) No 1107/2009 of the European Parliament and of the Council of 21 October 2009 concerning the placing of plant protection products on the market and repealing Council Directives 79/117/EEC and 91/414/EEC.

[g] Regulation (EC) No 1223/2009 of the European Parliament and of the Council of 30 November 2009 on cosmetic products.

[h] Commission Regulation (EU) No 1130/2011 of 11 November 2011 amending Annex III to Regulation (EC) No 1333/2008 of the European Parliament and of the Council on food additives by establishing a Union list of food additives approved for use in food additives, food enzymes, food flavourings and nutrients.

[I] Regulation (EC) No 1831/2003 of the European Parliament and of the Council of 22 September 2003 on additives for use in animal nutrition

[j] Regulation (EC) No 850/2004 of the European Parliament and of the Council of 29 April 2004 on persistent organic pollutants and amending Directive 79/117/EEC.

The European Chemical Agency (ECHA) is the driving force among regulatory authorities in implementing the EU chemical legislation covering the majority of registered substances that are on the EU market. However, some substances considered as CECs are regulated by other European agencies e.g. pharmaceutical compounds regulated by EMA.

2.2 Prioritisation of candidate substances

Once the relevant chemical categories are identified, it is important to prioritise the corresponding (or candidate) substances in order to select those of higher concern and to develop a manageable list of substances for the risk management of RWAI.

2.2.1 Identification of potential hazards and exposure

Currently, ECHA is performing an "integrated selection and priority setting" exercise using screening methods to identify 'Substances of Very High Concern' (SVHCs) in the EU using information from the REACH registration (22,023 unique substances by March 2019) and the CLP (147,549 substances notified by February 2019 and 4,264 with Harmonised Classification and Labelling by March 2019) databases as well as external information sources (e.g. scientific literature, online chemical databases). The objective of this screening is to identify which of the potentially hazardous substances have a high potential for exposure to humans or the environment by combining selected hazard data with use and exposure information as following:

- Persistency, Bioaccumulation and Toxicity properties (PBT/vPvB)
- Carcinogenicity, Mutagenicity, Reprotoxicity (CMR) cat 1A/1B
- Endocrine disruption (ED)
- Sensitisation
- High tonnage for wide dispersive uses

This exercise will contribute to implementation of the SVHC Roadmap, which aims to have all relevant currently known SVHCs included in a Candidate List by 2020 with a view to having full clarity on all registered substances by 2027. The output of this ECHA initiative will be highly relevant in the context of setting MQR for RWAI. However, it should also be noted that the following categories of chemicals, that are potentially relevant for reclaimed water reuse, are not included in the SVHC roadmap: medicinal product substances, food or feeding additives and unintentional by-products of industrial processes (except persistent organic pollutants; POPs). This is because these chemicals are not within ECHA’s scope due to REACH registration and CLP notification exemptions. Moreover, PBT/vPvB and ED effects are not currently a classification criterion under the CLP regulation, which currently focuses on environmental safety from the perspective of aquatic ecosystems only. Therefore, for the purpose of identifying and prioritising substances for RWAI, it will be necessary to complement the ECHA list with further prioritisation exercises on the relevant excluded chemical categories and selection criteria.

2.2.2 Declassification criteria: treatment steps and technologies at the urban wastewater treatment and reclamation plants

In a second tier, prioritised substances could be declassified based on the reported treatment efficacy of the specific substance at urban WWTPs and reclamation plants. Wastewater treatment processes are generally referred to as primary (physical process eliminating mainly visible material), secondary (biological process removing organic matter through the use of microorganisms), tertiary (chemical process removing nitrogen and phosphorus), disinfection (for removing pathogens) and advanced (removing micropollutants) processes. Substance/treatment-specific removal efficiencies should be estimated and reported for each prioritised substance. In principle, removal efficiencies are available for all substances identified within drinking water standards (e.g. EU Drinking Water Directive EU, 1998 and the World Health Organization drinking water standards WHO, 2017). Gorito et al (2017) review several studies on removal performances by constructed wetlands for 24 PSs, 2 other substances with EQS as well as 8 CECs on the watch list of substances pursuant to the EQS Directive (EU, 2008). For pharmaceuticals, substance/treatment specific removal efficiencies have been investigated as, for example, by primary and secondary clarifiers, bioreactors and sorption to primary sludge (Stasinakis et al., 2013) or by photocatalytic degradation (Paredes et al., 2019). Rizzo et al. (2019) critically reviewed the best available technologies for the advanced treatment of urban wastewater including consolidated (ozonation, activated carbon and membranes) and new advanced treatment methods (mainly advanced oxidation processes) analysing their efficiency in the removal of CECs. In addition, Krzeminski et al. (2019) discussed the performance of secondary wastewater treatments for the removal CECs that can be implicated in wastewater reuse practices.

3. Methodological approach to derive chemical quality standards for RWAI

3.1 Identification of the environmental compartments, populations and individuals at risk of direct or indirect exposure

The EC reclaimed water reuse regulation proposal (EU, 2018) requires that MS ensure that the use of RWAI has no adverse effects on environmental matrices: soil, groundwater, surface water, and dependent ecosystems, including crops to be irrigated. Indeed, both humans and other organisms in the environment can be exposed directly (i.e. receipt of irrigation water, splashes or spray drift) or indirectly (i.e. bioaccumulation within the food web) during water reclamation and agricultural irrigation operations. However, the direct exposure of workers and residents in the framework of operational safety is not addressed in this paper that focuses solely on environmental assessment (including humans via the environment).

Under the environmental assessment, both the soil and the water compartments can be exposed to reclaimed water releases at different levels according to the type of crops and farming practices adopted. The relevant soil compartment is the agricultural landscape and the pertinent water compartments are water bodies in the vicinity of the agricultural landscapes that could receive water releases during irrigation operation. Depending on the location of the agricultural landscapes, these water bodies include surface waters (i.e. rivers, lakes), coastal zones (i.e. marine water) and groundwater. As a consequence, the organisms to be protected (defined as protection targets) are those living in these ecosystems, directly exposed to reclaimed water but also the organisms that consume water and/or food (predatory organisms) from these ecosystems and can hence be indirectly exposed through the accumulation of the chemical contaminants in the food chain (secondary poisoning). The predatory organisms are defined as predators or top predators, depending if a simple or more complex food web is assumed.

For water bodies, the populations at risk considered in several EU regulations are pelagic organisms, benthic organisms, predators and top predators (for the marine food web) and the individuals at risk are humans via ingestion of drinking water and fishery products. For the soil compartment, the populations considered as at risk are the terrestrial communities (microorganism, invertebrates, plants), worm-eating predators (birds and mammals) and top predators of these ecosystems, and the individuals at risk are humans consuming animal products (meat, milk, eggs) and crops.

3.2 Evaluation of the applicability of the EQS derivation methodology under the WFD to the use of RWAI

The German Environment Agency (UBA, 2017) emphasised that quality requirements for reclaimed water reuse should comply with and complement the current EU legislation that already exists for surface water and groundwater protection, and in particular the WFD principles and resulting quality standards. The EC proposal for a water reuse regulation (EU, 2018) also states that, amongst others, the regulatory requirements of the WFD and its daughter EQS and PS Directives have to be fulfilled. Therefore, we believe that the relevance of using EQS for RWAI and the potential need for further adaptation must be investigated.

In 2005, a technical guidance document was prepared (Lepper, 2005) for the purpose of EQS derivation that comply with the requirements of Annex V of the WFD. It was further updated in 2011 to develop the steps required to derive EQS for metals, and EQS for biota and sediment (TGD, 2011). Recent developments specify the methodology for the use in the derivation of biota standards which address human health and secondary poisoning of wildlife, and the derivation of EQS for bioavailable metals (TGD, 2018). It is important to note, however, that the EQS are defined in relation to the protection of organisms from direct chemical exposure in surface waters and organisms from indirect exposure via the aquatic food chain (secondary poisoning) and that the use of RWAI is out of scope of EQS derivation. In order to assess if existing EQSs are relevant for this new purpose, the protection targets of the EQSs have been compared with those identified for RWAI. Practically, the methodology for EQS derivation involves the previous derivation of quality standards (QSs) for each identified protection target with a Maximum Acceptable Concentration (MAC) and an Annual Average (AA) concentration established in surface waters and AA concentrations established in biota and sediment. The protection targets of RWAI are listed in the Table 2 together with their relevant identified applicable QS from EQS derivation methodology. The absence of existing QS for some protection targets is also indicated.

Table 2: Protection targets to be addressed for the use of RWAI and existing relevant Quality Standards from the TGD reaching this goal

Protection target		**Related exposure source**	**Relevant QS of the TGD**
Freshwater ecosystem	Pelagic organisms	Freshwater	$MAC\text{-}QS_{fw,\ eco}$ $AA\text{-}QS_{fw,\ eco}$ (µg.L-1)
	Benthic organisms	Freshwater sediment	$QS_{sediment,\ fw}$ (µg.kg-1sed ww or dw)
	Predators (birds and mammals)	Freshwater prey (e.g. fish and molluscs)	$QS_{biota\ fw,\ sec\ pois}$ (µg.kg-1biota)
Marine ecosystem	Pelagic organisms	Saltwater	$MAC\text{-}QS_{sw,\ eco}$ $AA\text{-}QS_{sw,\ eco}$ (µg.L-1)
	Benthic organisms	Saltwater sediment	$QS_{sediment,\ sw}$

			(µg.kg-1sed ww or dw)
	Predators (birds and mammals)	Saltwater prey (e.g. fish and molluscs)	$QS_{biota\ sw,\ sec\ pois}$ (µg.kg-1biota)
	Top predators (e.g. killer whales and polar bears)	Birds and mammals	$QS_{biota\ sw,\ sec\ pois}$ (µg.kg-1biota)
Terrestrial ecosystem	Agricultural soil organisms incl. crops	Agricultural soil	No existing QS
	Predators (birds and mammals)	Terrestrial prey (e.g. earthworms and plants incl. crops)	$QS_{biota\ fw,\ sec\ pois}$ (µg.kg-1biota)
	Top predators (e.g. raptors and mustelids)	Small birds and mammals	No existing QS
Humans	Humans consuming fishery products	Freshwater and saltwater fish, molluscs…	$QS_{biota,\ hh\ food}$ (µg.kg-1biota)
	Humans consuming drinking water	Abstracted water and Groundwater	$QS_{dw,\ hh}$ (µg.L-1)
	Humans consuming irrigated crops and animal products	Crops Meat, milk and eggs	No existing QS

3.2.1 Protection targets covered by existing QSs

As expected, QSs derived to protect aquatic organisms, their predators/ top predators and humans during drinking water and fishery products consumption under the WFD Common Implementation Strategy will also be protective for the use of RWAI. More surprising, the QS for biota protecting predators consuming freshwater fish ($QS_{biota\ fw,\ sec\ pois}$) and derived according to the diet-based concentration method of the TGD (2011) could also be considered protective for terrestrial predators like birds and mammals eating worms and/ or plants because the toxicological data on birds and mammals can be considered also relevant for terrestrial predators. However, this methodology makes no difference between various food items and use a default factor of 3 to correct for the differences in caloric content between standard laboratory food on the one hand and prey species in the field on the other hand. According to Verbruggen (2014) this default factor is reasonable for fish (factor 2.8) but is not for earthworms (factor 5.2), which a have a much lower caloric content based on fresh weight. It is probably also the case for plants meaning that $QS_{biota\ fw,\ sec\ pois}$ could be insufficiently protective for terrestrial worm-eating and plant-eating predators.

3.2.2 Non-covered protection targets by existing QSs

Other organisms exposed via the soil (by direct or indirect exposure to reclaimed water) are not covered by an existing QS:

- The terrestrial organisms including microorganisms, invertebrates, plants via direct exposure and top predators consuming small birds and mammals via indirect exposure through bioaccumulation in the food chain.
- Humans via indirect exposure through bioaccumulation in the food chain (crops and animal products).

3.3 Proposal for the adaptation of existing QS or the development of new QS for RWAI use:

For pelagic organisms (and benthic organisms if the Equilibrium Partitioning Method is used), the QSs of the WFD can be considered over-protective because reclaimed water will be diluted within receiving water bodies. Increasing the QSs concentration by the factor of dilution of receiving freshwater and marine water bodies as normally considered in chemical risk assessment could be implemented for RWAI.

For terrestrial organisms including microorganisms, invertebrates and plants, the methodology to derive Predicated Non-Effect Concentration for the soil (PNECsoil), in different EU regulatory frames is recommended to develop a new QS_{soil} (ECHA, 2008). However, it should be noted that terrestrial data are available only rarely for many chemicals and the equilibrium partitioning method commonly used in case of terrestrial data lacking is driving a high uncertainty on the estimated PNECsoil. Moreover, the actual scoping of PNECsoil derivation does not include terrestrial invertebrates living above-ground (e.g. ground dwelling beetles), terrestrial vertebrates living a part of their lifetime in soils (e.g. mice) and groundwater organism (invertebrates and micro-organism).

For terrestrial predators (birds and mammals) and top predators like raptors and mustelids consuming small birds and mammals, the caloric content-based diet concentration methodology described by Verbruggen (2014) can be adopted when both the energy content and bioaccumulation parameters are available for several food items in order to select the critical food item in the food chain that is most relevant for secondary poisoning in agricultural soils.

For humans exposed through the terrestrial food chain, the existing general food standards established for crops and animal products for relevant population groups could be used as a first instance. For example, the WHO Codex Alimentarius (including food additives, residues of pesticides, veterinary drugs and contaminants), the EC Maximum Residues Levels for pesticides in food (EU, 2005) and the EC maximum levels for certain contaminants in foodstuffs defined for selected metals, PCBs, dioxins and polycyclic aromatic hydrocarbons

(EC, 2006) could be drawn upon. If such food standards are not available for the substances and/or food of concern, the methodology used to derive the QS intended to protect humans against adverse health effects from consuming contaminated fishery products ($QS_{biota, hh food}$) in the TGD (2018) could be adapted considering all the various sources of food consumption of humans (fish, crops and animal products).

4. Data gaps, uncertainties and research needs

4.1 Chemical contaminants in reclaimed water: transformation products and unregulated substances

Urban WWTPs and water reclamation plants are designed to remove chemical substances via various biotic and abiotic degradation processes. Therefore, transformation products, also referred to as metabolites, are expected to be generated at treatment plant facilities. For example, Paredes et al. (2019) demonstrated that in a secondary wastewater effluent resulting from photocatalytic degradation, 156 transformation products originating from eight pharmaceuticals could be detected. However, in most of the studies designed to assess wastewater treatment removal efficiency, transformation products are not identified as the main focus is on the disappearance of the parent substance. It is an important issue since it has been reported that transformation products can sometimes represent a higher toxicity concern than parent substances. For example, carbamazepine-10,11-epoxide, a main metabolite of the antiepileptic drug carbamazepine, is reported to exhibit a higher chronic toxicity on the midge *Chironomus riparius* in comparison to the parent compound (Heye et al., 2016). Therefore, it is highly recommended that future investigations include the identification and quantification of transformation products and assess their toxicity in complementary studies. Another uncertainty is related to the occurrence of unregulated "substances" including those transported in storm water runoff, illicit substances etc., that could reach municipal WWTPs as, for example, reported in the Netherlands and Spain by Bijlsma et al. (2012 and 2014) and, more recently, microplastics that are not removed by WWTP (Lares et al., 2018) and persist in the environment (De Souza Machado et al., 2018). Therefore, a better characterisation of the impact of storm water run-off and microplastics, as well as the occurrence of illicit substances in urban WWTPs, is required.

4.2 Prioritisation and QS derivation methodology: Data on terrestrial ecosystems and QS biota conversion

Soil is a primary exposed environmental compartment within RWAI and, whilst exposure after sludge application is considered within regulatory chemical risk assessment, the use of RWAI is not. Data recorded over many years of RWAI practice in the Tula Valley (Mexico) show a mean retention of 86% of chemical contaminants in soil (Navarro et al., 2015) and Carter et al. (2019) recently reported that RWAI continuous application has resulted in pharmaceuticals building up in soils to total concentrations of up to ca. 15 mg kg^{-1}. However, regulatory data on soil are mostly missing. Indeed, until recently, data on freshwater aquatic ecosystems were considered the minimum necessary dataset under most regulations. Biocides and PPPs regulations are an exception, requiring the submission of terrestrial data on corresponding active substances. In order to perform a targeted prioritisation exercise of contaminants in RWAI and derive sound QS for the soil communities, it is necessary to investigate their terrestrial persistence, bioaccumulation through the food chain and toxicity including endocrine disrupting effects.

Finally, the EC methodology to derive EQS supports the conversion of QSs developed for biota in the aquatic food web into equivalent aquatic concentrations in order to allow monitoring in water only. For the conversion of a terrestrial biota standard into a soil concentration, a similar calculation can be made using the relevant biota-to-soil-accumulation-factor (BSAF, usually for earthworms) and biomagnification factors from preys to predators (usually from earthworm to small terrestrial birds or mammals) as suggested by Verbruggen (2014). In order to use a similar methodology for humans consuming irrigated crops and animal products, it would be necessary to develop BSAFs in the different irrigated crops and bio transfer factors (BTF) in animal products (e.g. milk, meat, eggs). Finally, a conversion of soil concentration into water concentration would require the development of models which estimate the soil concentration resulting from the repeated application of RWAI to fields, as currently undertaken for sludge applications in most of the chemical regulations (e.g. REACH, BPR, medicinal products for human use).

5. Conclusions

A barrier to the greater use of RWAI is the perceived risks the practice may pose to human health and the environment, especially related to CECs identified as not fully removed within urban WWTPs or reclamation plants. This analysis has shown that current legislation pertaining to industrial releases, WWTPs and environmental protection is not sufficient to manage the risk posed by hazardous substances within a reclaimed water reuse context. This is understood to be a function of the fact that existing legislations were not specifically developed to address reclaimed water reuse pathways and protection targets. Thereby, almost all chemicals categories intentionally manufactured or the by-products of industrial processes could reach urban WWTP. The vast majority of these chemicals being regulated under the authority of the European agencies, it is therefore highly recommended to use the output of their prioritisation methods, such as the SVHC roadmap by ECHA, in order to identify and prioritise the substances of concern for RWAI. Special attention, however, should also be given to excluded chemical categories (i.e. substances out of scope from a European agency perspective) when undertaking the specific prioritisation exercise together with consideration of the relevance of prioritisation criteria for the reuse of RWAI. The declassification of substances from the priority list should be made possible based on data on removal efficiencies of wastewater treatment technologies. Once a list of priority substances has been defined, QS can be derived in order to monitor the chemical quality of reclaimed water and ensure compliance with human health and environmental protection goals. Under the EU WFD, a methodology is available to derive EQS for contaminants in order to protect water bodies from chemical pollution. In the present analysis we conclude that while the same methodology is appropriate with regard to protection of the water ecosystems (and their consumers) from run-off during irrigation, the main exposed organisms in soil or via the terrestrial food chain would not be adequately protected under this approach. In parallel with ongoing research, further work will draw on knowledge already available under different regulatory frameworks to develop such a methodology including, for example, opportunities to adapt current QSs or derive new QSs for terrestrial protection targets as required. Overall, this work provides guidance to policy makers, researchers and practitioners with regard to both the need and opportunities for the derivation of QS for CECs in RWAI. Key data gaps have been identified concerning soil ecosystems, wastewater treatment technology removal efficiencies and the occurrence, behaviour and fate of transformation products generated during the reclaimed water reuse cycle requiring further investigations at both research and regulatory levels. As a future perspective, we believe that in order to facilitate moves towards managing the risks posed by chemicals in the context of RWAI, standardised emission scenarios should be developed for this use, supporting systematic assessment of risks posed by regulated chemical substances released from WWTPs.

Conflicts of interest

The authors have no conflict of interest to declare regarding this article.

Acknowledgements

The authors thank Dr Joze Roth for providing reviews on the sources of wastewater to UWWTP and European policy landscape. The authors would like to acknowledge the financial support provided by COST-European Cooperation in Science and Technology, to the COST Action ES1403: New and emerging challenges and opportunities in wastewater reuse (NEREUS). Disclaimer: The content of this article is the authors' responsibility and neither COST nor any person acting on its behalf is responsible for the use, which might be made of the information contained in it.

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