THE UNIVERSITY OF ARIZONA
Cooperative Extension

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Plant Uptake of Contaminants of Emerging Concern During Irrigation with Recycled Water

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What is recycled water?

Recycled (or reclaimed) water is municipal wastewater treated to a sufficient quality for its intended purpose—a concept known as 'fit-for-purpose' reuse. In any region with impaired or scarce water resources, recycled water can be used to replace or augment existing water supplies. Potential applications range from power plant cooling to drinking water augmentation to agricultural irrigation. While the U.S. is considered a leader in potable reuse—the use of advanced treated recycled water as a drinking water supply—Israel has established itself as a leader in agricultural reuse. In fact, Israel now recycles 75% of its wastewater for use in agricultural applications (CONSERVE, 2019). This dramatically reduces the country's reliance on scarce freshwater resources or costly water supply alternatives such as desalination. With global food demand expected to increase 70% by 2050 (UNESCO, 2012) and climate change expected to put further strain on freshwater supplies, agricultural reuse is slated for further expansion in the future.

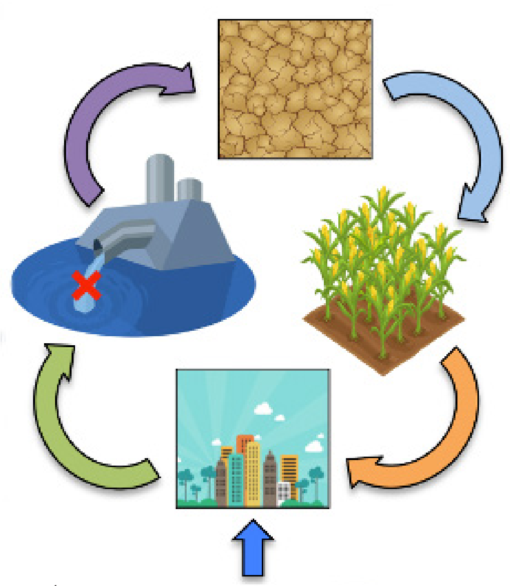


Figure 1.

Use of recycled water for irrigation in the U.S.

According to the U.S. Geological Survey, freshwater withdrawals in the U.S. totaled 280 billion gallons per day in 2015, with irrigation accounting for 118 billion gallons per day (or 42% of the total) (USGS, 2018). Of the ~34 billion gallons per day of wastewater generated in the U.S., only 0.7 billion gallons per day were recycled for irrigation uses in 2015. This represents a 42% increase since 2010, with more than 40 states now participating (Thebo, 2017), but it still only represents 0.6% of the total water used for irrigation.

Theoretically, more agricultural reuse could further reduce freshwater demand, but there are a number of practical and institutional barriers limiting broader adoption of this practice. In many places, competition between sectors (e.g., potable vs. agricultural vs. industrial) may limit the amount of recycled water available for a specific application. In other locations, the distance between major supply centers (i.e., urban areas generating wastewater) and agricultural fields (rural communities) may render agricultural reuse economically or technically infeasible due to the high cost of bulk water transmission. The 35 largest sites with 'untapped' agricultural reuse potential, meaning the reuse source is in close proximity to the agricultural demand, could provide an additional 1 billion gallons per day of water for 200,000 acres of cropland (Thebo, 2017). This would still account for only a small fraction of overall irrigation demand, but it would more than double the current reuse total for irrigation and might be critically important on a local scale in water-stressed regions.

Another barrier to agricultural reuse relates to the implications of water quality for public perception and public health protection. Depending on the type of treatment, recycled water may contain elevated concentrations of nitrogen and phosphorus, thereby reducing the need for fertilizer. However, recycled water may also contain a number of constituents of emerging concern (CECs). CECs are broadly defined as "any synthetic or naturally occurring chemical or microorganism that is not commonly monitored in the environment but has the potential to enter the environment and cause known or

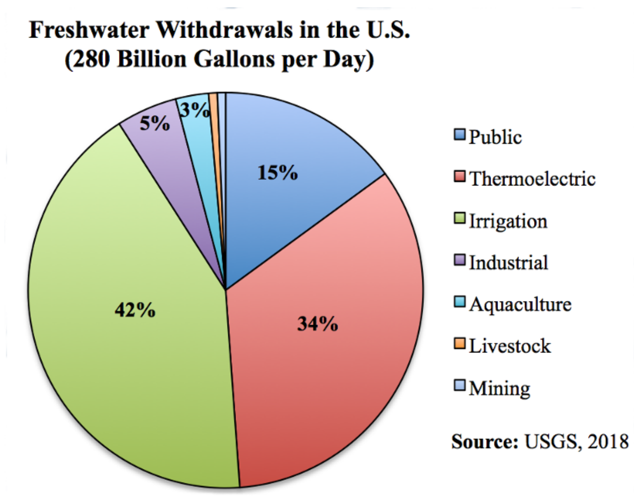


Figure 2.

suspected adverse ecological and/or human health effects" (USGS, 2017). CECs include pharmaceuticals and personal care products (PPCPs), per- and polyfluoroalkyl substances (PFAS), and even antibiotic resistant bacteria (ARB) and antibiotic resistance genes (ARGs), all of which have been detected in recycled water (Michael et al., 2013; Rizzo et al., 2013; Bacaro et al., 2019). It should be noted that these compounds have also been detected in surface water and even in treated drinking water, and thus the issue is not limited to recycled water (Glassmeyer et al., 2017). The Food Safety Modernization Act (FSMA), while currently in abeyance, addresses some microbial concerns, specifically limiting the geometric mean *E. coli* concentration in agricultural irrigation water to ≤126 CFU/100 mL. Existing guidelines and/or regulations for microorganisms in recycled water are generally even more stringent, often requiring non-detects for total and/or fecal coliform bacteria. However, regulations for agricultural applications do not currently address broader occurrence of CECs.

What are CECs?

PPCPs include a diverse suite of chemical substances, including over-the-counter and prescription drugs for human and veterinary uses, and other products that are used by individuals for personal health or cosmetic purposes. PPCP concentrations in treated wastewater vary by orders of magnitude depending on inputs to the local collection system and the unit processes at the wastewater treatment facility. Concentrations in recycled water generally range from the low ng/L (part-per-trillion) to µg/L (part-per-billion) level. Importantly, the general consensus is that typical concentrations of the vast majority of PPCPs in recycled water pose de minimis risks to public health.

PFAS represent a potential exception in that recently published health advisory levels are in the low ng/L range. Specifically, the U.S. Environmental Protection Agency (EPA) published a combined health advisory level of 70 ng/L for perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), while California recently lowered its individual notification levels for PFOA and PFOS to 5.1 ng/L and 6.5 ng/L, respectively. PFAS are a family of molecules composed of a carbon backbone that is fully or partially fluorinated, and the number of carbon atoms in the backbone is a defining characteristic. For example, PFOA and PFOS both contain 8 carbon atoms within their backbone, hence their 'octan-' designation. The C-F combination represents one of the strongest bonds in organic chemistry and ultimately makes the compound highly persistent through water and wastewater treatment and in the environment. Water and food consumption are generally considered two major routes of human PFAS exposure. Additional information related to PFAS can be found in Dery et al. (2019).

Although exposure to PPCPs in recycled water is unlikely to cause any adverse public health impacts, there may be ancillary effects of certain subgroups, specifically antibiotics. The main concern for the release of antibiotics into the environment is related to the proliferation of antimicrobial resistance, either as intact ARB or as intracellular or extracellular ('free') ARGs that can be transferred to pathogenic bacteria via horizontal gene transfer. In fact, wastewater treatment plants have been identified as 'hotspots' for the spread of ARB and ARGs into the environment (Rizzo et al., 2013). Although clinical antimicrobial resistance has become a clear threat to public health, the role of treated wastewater is still unclear

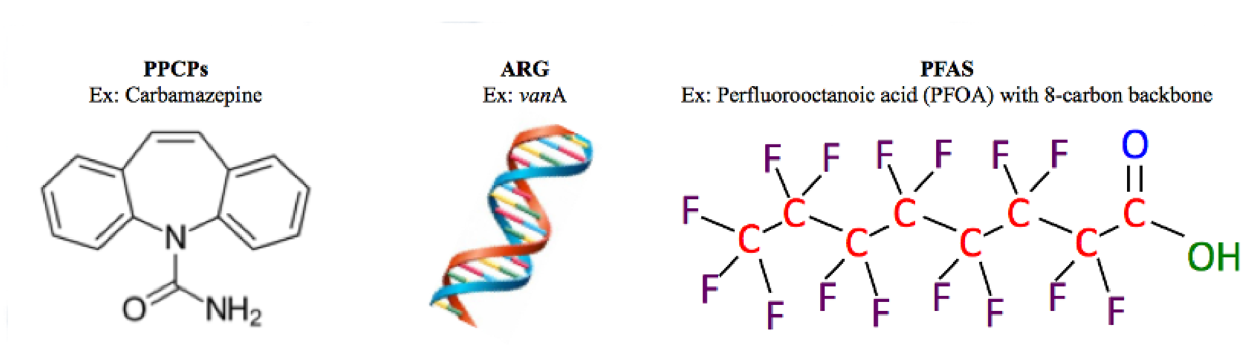
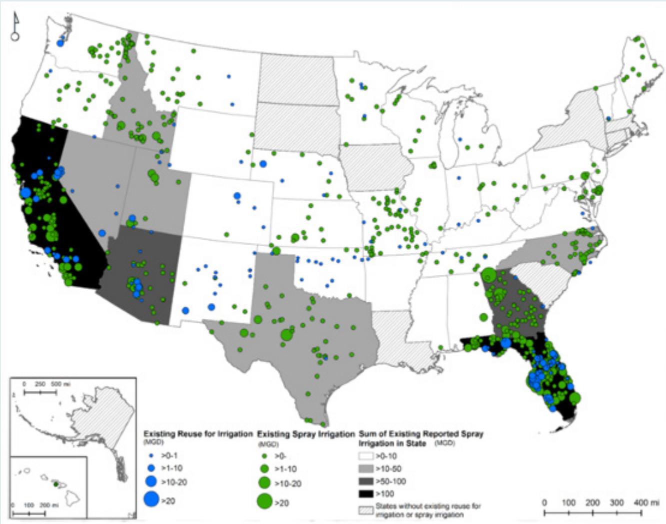
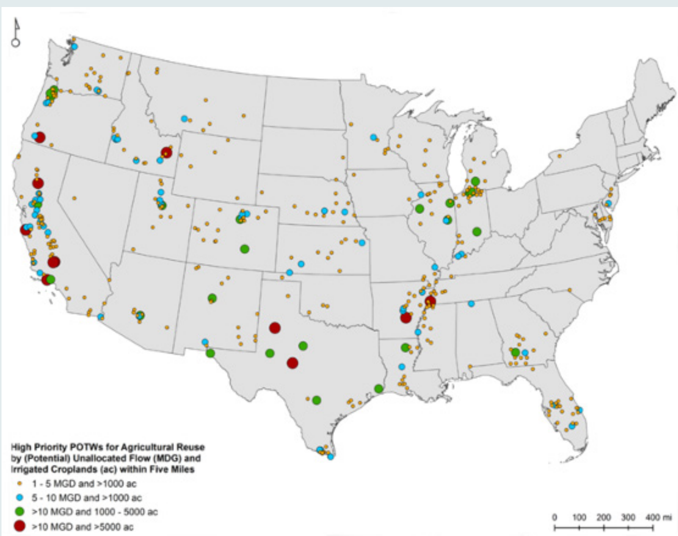


Figure 3.





Top: Existing implementation of recycled water for irrigation applications in the U.S.

Middle: Publicly owned treatment works (POTWs) identified as having significant recycled water flows available for nearby croplands.

Bottom: States permitting recycled water use for food and non-food crops, non-food crops only, or neither.

Take-home message: Geospatial disconnects (i.e., proximity between supply and demand for recycled water) can be a significant barrier to agricultural reuse. In other words, it may not be practical to transport recycled water from large urban areas to distant agricultural fields. However, studies have identified numerous facilities with agricultural reuse potential.

Source: Anne Thebo of the Pacific Institute, 2017

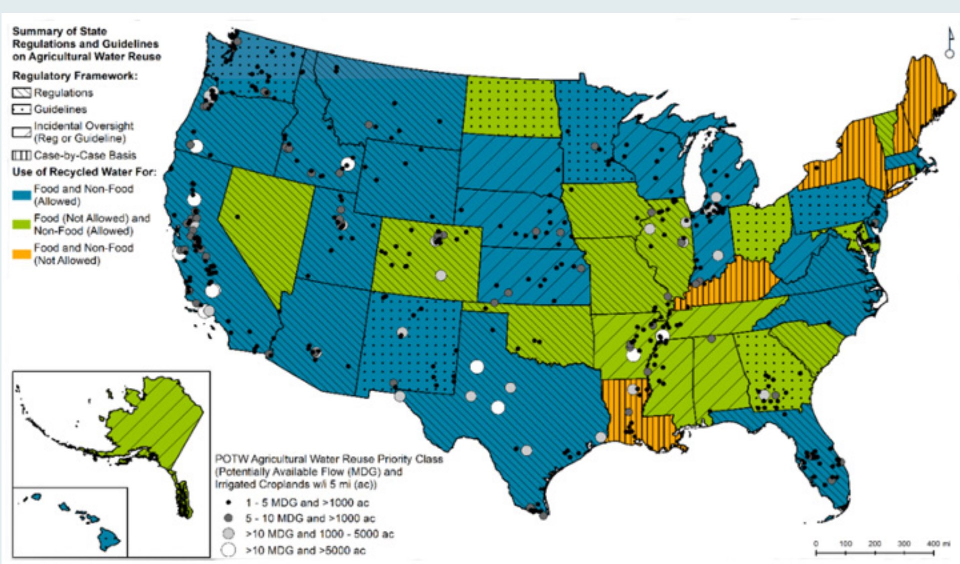


Figure 4.

Why are CECs relevant to agricultural reuse?

CECs may enter agroecosystems through irrigation with recycled water or through land application of biosolids (organic matter recycled from sewage, commonly used in agriculture). Upon irrigation, CECs may be deposited onto the plant tissue itself or may percolate through the soil, potentially being transported into the roots and then throughout the internal structure of the plant (Christou et al., 2017; 2019a). The potential for plant uptake of CECs is highly complex and depends on myriad factors involving the soil, water, plant (and location within the plant), and the CEC in question.

CEC uptake by plants is often described on a mass per dry weight basis. For example, the antibiotic sulfamethoxazole has been detected at ~5 ng/g of tomato, and the antibiotic ciprofloxacin has been detected at ~10 ng/g of cabbage or carrot (Christou et al., 2017). Summed PFAS concentrations sometimes exceed 1 μg/g in edible plant tissue, although this is generally observed only when CECs are spiked at high concentrations or in the presence of biosolids (Ghisi et al., 2019). Bioconcentration factors (BCFs), or the ratio of plant tissue concentration to soil concentration, have also been used, with BCFs >1.0 indicating a high potential for uptake. For example, BCFs for PFOA and PFOS (8-carbon compounds) are often less than 1.0 to 2.0, with most of the compound detected in plant roots. On the other hand, 4-carbon PFAS compounds can have BCFs higher than 100, with detection shifting to edible portions of plants (Ghisi et al., 2019). This occurs because shorter-chain compounds preferentially partition to water, thereby increasing their mobility into plant tissues, whereas longer chain compounds adsorb more readily to organic matter in soil.

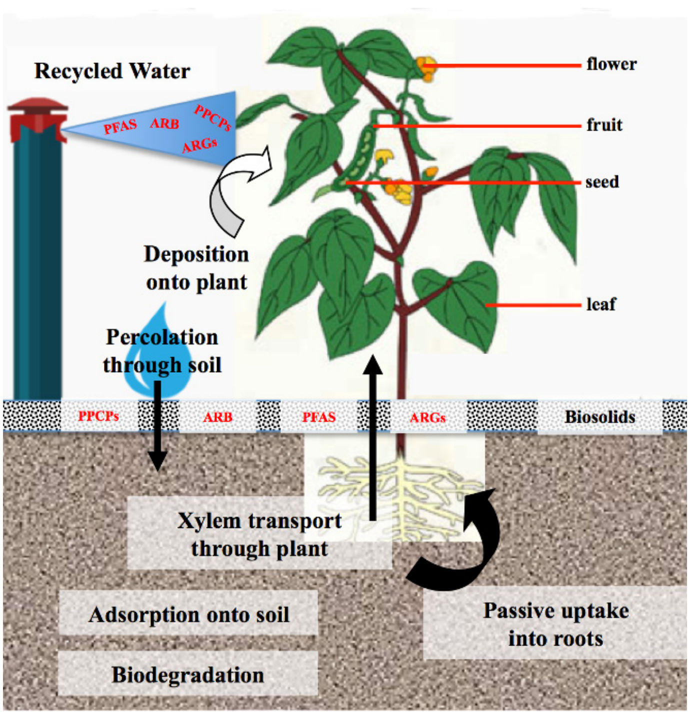
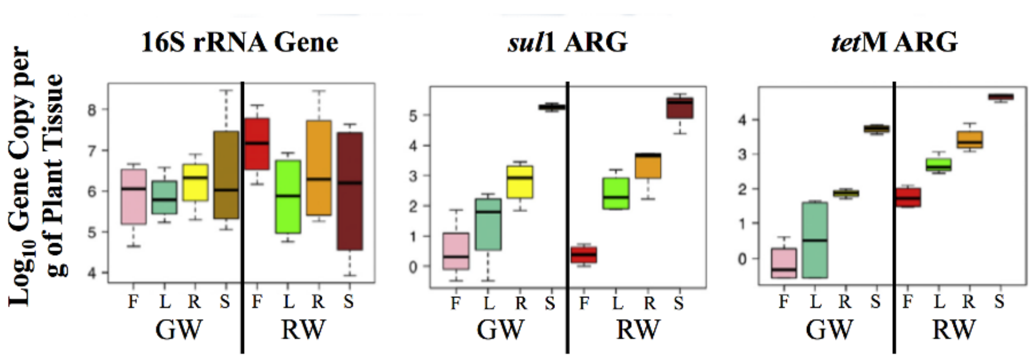


Figure 5.

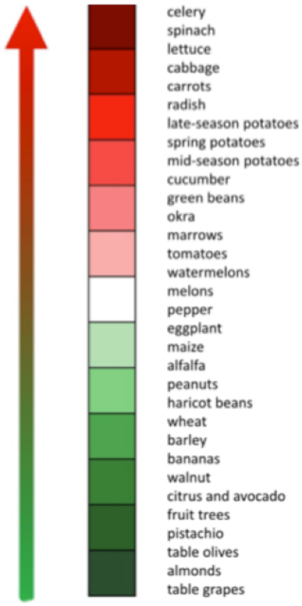
With respect to ARB and ARGs, some studies indicate that irrigation with recycled water has no discernible impact on antimicrobial resistance within the soil microbiome (Negreanu et al., 2012), while other studies have demonstrated higher prevalence of certain ARGs in soils irrigated with recycled water (Wang et al., 2014; Han et al., 2016). The figure below highlights these contradictory observations. For context, the 16S rRNA gene is often used as a surrogate for bacterial abundance in a sample, and sul1 and tetM are ARGs conferring resistance to the antibiotics sulfamethoxazole and tetracycline, respectively. Based on the figure, irrigation with groundwater vs. recycled water had minimal impact on abundance of the 16S rRNA gene, or overall bacterial abundance. There was also little impact on the sul1 ARG, but there was a clear increase in tetM with recycled water. For both sul1 and tetM, ARG occurrence consistently decreased from soil to roots to leaves to fruit—often by several orders of magnitude (Cerqueira et al., 2019a; 2019b).

Christou et al. (2019a) summarizes various biotic and abiotic factors affecting CEC uptake by plants. These factors include target CEC characteristics, plant type, soil type, and environmental conditions, including conditions that might promote CEC biodegradation within the soil. With respect to plant type, experimental results suggest CEC uptake is greatest in leafy vegetables; followed by root vegetables, cereals, and fodder crops; and least in fruits and nuts.



Notes: F = Fruit L = Leaf R = Root S = Soil GW = Groundwater RW = Recycled Water
Source: Cerqueira et al. (2019)

Figure 6.



Conditions Contributing to CEC Uptake
High evapotranspiration rate
High net irrigation rate
Healthy (non-stressed) plants
Growing season during summer
High temperature
High wind speed
Low humidity
Adequate soil moisture
Acidic pH
Aerobic conditions
Low soil organic matter content
Sandy (vs. clay/loamy) soils
Low lipid content in roots

Source: Christou et al. (2019)

Figure 7.

What does this mean for environmental and public health?

Laboratory experiments have shown that CECs can affect the development and physiology of plants, but these experiments often involve soil-free hydroponics systems with high CEC concentrations in the water (Christou et al., 2019b). In general, the scientific literature suggests there is potential for CECs to adversely impact crop health and for antimicrobial resistance elements to be transferred to pathogenic bacteria in the environment, but these outcomes have not yet been demonstrated conclusively under realistic environmental conditions.

With respect to public health, Prosser and Sibley (2015) calculated hazard quotients for PPCPs based on plant uptake data reported in the literature. A hazard quotient is the ratio of exposure to some toxicological benchmark. That benchmark might be a 'no adverse effect level' or sometimes may reference a therapeutic dose, but it often includes a large safety factor to account for additional toxicological uncertainty. It is assumed that hazard quotients less than 1.0 represent a safe level of exposure to the constituent in question. Prosser and Sibley (2015) found that for studies involving recycled water with environmentally relevant CEC concentrations, the highest hazard quotient for adults was 0.08 (for a veterinary nonsteroidal anti-inflammatory drug), and most hazard quotients were significantly lower. For toddlers, five compounds had hazard quotients greater than 0.1, but none were greater than 0.4. The study concluded that individual PPCPs detected in edible tissues of plants represented a de minimis (insignificant) risk to human health. However, they cautioned that potential public health implications of complex PPCP mixtures were still unclear. Also, more data are needed for PFAS uptake with environmentally relevant concentrations and in the absence of biosolids to better characterize the public health implications of using recycled water alone. PFAS are relevant to public health at lower concentrations than many PPCPs, which may result in higher hazard quotients, but additional data are needed for confirmation.

Even if hazard quotients suggest public health risks are negligible, there may be other ancillary effects of CEC exposure. In a study conducted in Israel, healthy people consuming produce irrigated with recycled water excreted higher urinary levels of the antiepileptic compound carbamazepine than healthy people consuming produce irrigated with freshwater (Paltiel et al., 2016). However, peak urinary levels were 10,000 times lower than the level expected after ingestion of a single therapeutic dose of carbamazepine. This offers further evidence that human exposure to CECs from produce irrigated with recycled water is possible but that the effects are likely insignificant, albeit detectable in some cases.

Quantitative microbial risk assessment (QMRA) is a common approach for estimating public health risks due

Example: Exposure to the Antibiotic Sulfamethoxazole in Carrots Irrigated with Recycled Water

Lowest Therapeutic Dose = 400 mg/day

Safety Factor ↓

Acceptable Daily Intake = (400 mg/day) / (70 kg body mass) / (1000) = 0.0057 mg/kg-day = 5700 ng/kg-day

$$\text{Estimated Daily Intake (ng/kg-day)} = \frac{\text{Concentration in Food (ng/g)} \times \text{Food Intake (cups/day)} \times \beta_1 \text{ (g/cup)} \times \beta_2 \text{ (unitless)}}{\text{Body Mass (kg)}}$$

Highest Observed Sulfamethoxazole Concentration in Carrots = 0.25 ng/g (dry weight)

Daily Carrot Intake = 2.80 cups/day (95^{th} Percentile)

β_1 = Conversion from cups to grams = 134.9 wet g/cup | β_2 = Conversion from wet weight to dry weight = 0.085 (U.S. EPA)

Adult: Body Mass = 76.6 kg | Estimated Daily Intake = 0.10 ng/kg | Hazard Quotient = 0.000018

Toddler: Body Mass = 15.4 kg | Estimated Daily Intake = 0.52 ng/kg | Hazard Quotient = 0.000091

Source: Prosser and Sibley (2015)

Approximately **5-20 tons of carrots per day** to reach acceptable daily intake

Figure 8.

to exposure to pathogenic microorganisms. QMRA is increasingly being used to evaluate the safety of potable reuse applications and to inform the development of regulations and design criteria for engineered treatment systems. These existing studies are possible because the data needed to perform a QMRA for waterborne enteric pathogens are often readily available. Theoretically, QMRA can also be applied to ARB and ARGs, but existing frameworks cannot be applied directly. The scientific community and water industry are currently collaborating to develop QMRA frameworks specifically to address antimicrobial resistance. Until that time, it is not possible to quantify the potential risks associated with antimicrobial resistance elements that might be detected in agroecosystems.

What does this mean for the use of recycled water in agriculture?

As demonstrated by many scientific studies, the concentrations of a large number of CECs are dramatically reduced during wastewater treatment. However, some CECs are recalcitrant (resistant to degradation) to conventional and even advanced water/wastewater treatment processes so they are likely to be detected in recycled water used for irrigation, with the exact compounds and concentrations being site-specific. As noted earlier, this is potentially problematic because a growing body of literature suggests that CECs can be taken up into plants irrigated with recycled water. When plants are exposed to environmentally relevant CEC concentrations, uptake is generally minimal, and studies suggest that the associated risks are negligible. However, growers and irrigation districts should be aware of this issue because perceived risks can still have a significant impact on the decision-making process. To address the remaining uncertainty, the scientific community is working closely with the water/wastewater and agricultural industries to better understand and characterize the implications for CECs in agroecosystems exposed to recycled water.

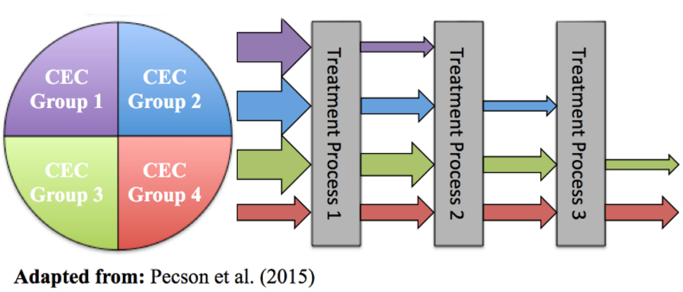


Figure 9.

For the latest information on this and related topics, please refer to the Center of Excellence at the Nexus of Sustainable Water Reuse, Food, & Health **(CONSERVE) at www.conservewaterforfood.org**

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