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Developing circular agricultural production systems

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Safe and sustainable use of bio-based fertilizers in agricultural production systems

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1 Introduction

Development of circular agricultural production systems necessitates the reuse/recycling/upcycling of agricultural byproducts (e.g. animal manure, crop residues, food waste and food processing waste) back into the production cycle thereby lessening the overall footprint of agriculture. Historically, many agricultural byproducts have been classified as 'wastes', as they were not the primary product of production. However, the goal of circularity is to utilize these 'wastes' as a resource instead of simply discarding them. Many agricultural byproducts contain significant amounts of nitrogen (N), phosphorus (P) and potassium (K) as well as a wide variety of micronutrients which makes them an excellent nutrient source for crop growth. Because of this, the development of bio-based fertilizers from a variety of agricultural byproducts is of increasing interest. In addition to supplying nutrients, bio-based fertilizers can improve soil health via the addition of organic carbon (C) which may improve soil structure, water holding capacity and water infiltration and support a healthy microbiome. Although these products contain valuable N and P for agricultural production, the conversion efficiency into edible food or other end products

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tends to be low with typical use efficiencies of 30% or less (Bordirsky et al., 2012; UNEP and WHRC, 2007; FAO, 2006) leading to potential environmental losses that impact air and water quality as well contribute greenhouse gasses (GHG) to the atmosphere which are associated with climate change. Efficiently integrating these bio-based fertilizers back into agricultural production is dependent on the characteristics of the product, availability, quality control, transportation costs/logistics, environmental regulation and public acceptance (Westerman and Bicudo, 2005). Additionally, valorization of these products is challenging as the nutrient contents tend to be diluted compared to synthetic fertilizer, less consistent and in some cases harder to handle and apply than traditional fertilizers. However, the additional benefits of providing a wider variety of micronutrients, compared to synthetic fertilizer, along with additions of organic C that may improve overall soil health and productivity, need to be taken into consideration as well.

In addition to concerns related to nutrient pollution, other environmental contaminants that may be associated with bio-based fertilizers include heavy metals, pathogens, antibiotics and chemicals of emerging concern (CEC; e.g. pesticides, hormones, polychlorinated biphenyls [PCBs], polycyclic aromatic hydrocarbons [PAHs] and microplastics; Tran et al., 2018; O'Connor et al., 2022; Tian et al., 2022). Bio-based fertilizers originating from biosolids, manures and other organic wastes can contain heavy metals (O'Connor et al., 2021). Heavy metals are toxic in high concentrations and are persistent pollutants in the environment where they can bioaccumulate and biomagnify through the food chain (Sardar et al., 2013). Pathogens may be present in many organic byproducts (livestock manure, manure compost, biosolids) used as bio-based fertilizers. The application of these fertilizers to agricultural crops, in particular those grown for fresh produce, has resulted in foodborne illness outbreaks (Alegbeleye et al., 2018; Callejón et al., 2015). The use of antibiotics in animal production and the presence of antibiotics in biosolids have raised concerns about the release of these drugs into the environment, particularly with land application of these byproducts, and the increasing prevalence of antibiotic resistant bacteria (ARB) and antibiotic resistance genes (ARGs) in the environment. Pesticide residues are commonly found in food wastes which may pose a threat to the quality of composts and digestates generated from these byproducts (Nguyen et al., 2020). The persistence and phytotoxic effects of these residues are unknown but may cause damage to soil quality and plant growth when applied to cropland as bio-based fertilizers at high rates (Boudh and Singh, 2019). There are a host of other CEC that are introduced into agricultural production and they remain persistent within the system. These chemicals have the potential to damage soil health and can pose health risks to both animals and humans. Given the large variety of agricultural byproducts that can serve as bio-based

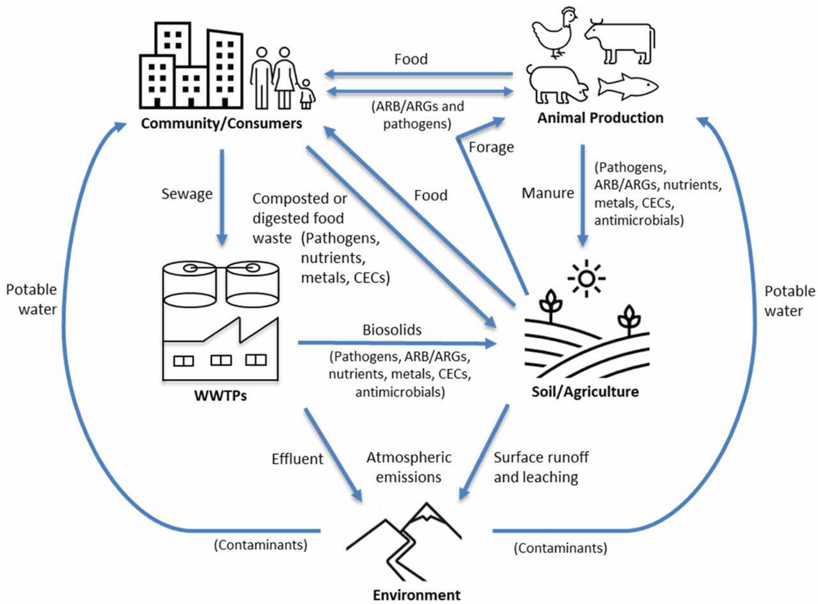

Figure 1 The routes by which biological and chemical contaminants in bio-based fertilizers are spread in environmental ecosystems. The contaminants make their way into the environment when manure, food waste and biosolids are applied to soils, either through direct application, animal excretion or disposal. The contaminants can then be transported in surface runoff or by leaching to surface and ground water, respectively, or emitted to the atmosphere. Humans and animals can be continuously exposed to contaminants via ingestion of food and water, while humans can also be exposed via contact in recreational waters.

fertilizers available for recycling within the agricultural system, it is important not only to consider the benefits in terms of C and nutrients, but also to assess any potential tradeoffs with respect to contaminants. Figure 1 illustrates some of the potential pathways of contaminant flow within circular systems.

2 Risk factors associated with utilizing bio-based fertilizers in agricultural production

2.1 Nutrient losses to the environment

Estimates of the conversion of N used in food production to that consumed by humans range from 10% to 20% (Bordirsky et al., 2012; UNEP and WHRC, 2007), with greater than 50% of N applied to cropland lost to downstream and downwind environments (Davidson et al., 2012). It is estimated that only 15% to 30% of applied phosphorus (P) is taken up by harvested crops (FAO, 2006). With these low nutrient use efficiency rates, the integration of agricultural byproducts as bio-based fertilizers into agricultural production needs to be carefully managed to reduce nutrient losses to the environment.

In terms of nutrients generated, livestock manure is one of the largest sources of bio-based fertilizers available. Globally, it is estimated that livestock production generates 131 and 23 Tg of manure N and P, respectively, per year (Liu et al., 2017; Zhang, 2017). Manure nutrient production is expected to increase as human and livestock populations increase and the dietary intake of meat increases in developing countries (Herrero and Thornton, 2013). While the quantity of manure nutrients generated over time are increasing, the growing disconnect between livestock and crop production on the agricultural landscape has left most modern livestock producers with a surplus of on-farm nutrients which contribute to environmental degradation in many regions. Overall, the greatest concerns have been related to air quality degradation due to emissions of NH_3, water quality impacts from nitrate (NO_3) leaching and P losses via runoff and climate impacts and ozone depletion from nitrous oxide (N_2O) emissions (Holly et al., 2018).

2.1.1 Reactive nitrogen losses to the environment

Reactive N (N_r) is essential to the growth of plants and animals and is typically the most limiting nutrient in agricultural production. The current rate of N_r loss to the environment, due to agricultural production, is more than 10 times the rate that occurred at the end of the 1800s (UNEP and WHRC, 2007) largely due to industrial-scale plant fertilizer production using the Haber-Bosch process in the twentieth century. Nitrogen losses are of increasing concern due to their negative consequences on human, animal and environmental health. The primary forms of N_r loss are ammonia (NH_3), nitrate (NO_3), nitrous oxide (N_2O) and N oxides (NO and NO_2 or NO_x).

Food production, including cropland and livestock production, is estimated to generate up to 90% of global NH_3 emissions (Ma et al., 2021; Zhan et al., 2021). Liu et al. (2022) estimated that, in 2010, approximately 58 Tg of N was lost as NH_3, with approximately 67% of that being generated from livestock production and land application of manures. Ammonia in the atmosphere is an important driver for the formation of fine particulate matter ($PM_{2.5}$) which is both an air quality and human health concern. In addition, the deposition of this N into terrestrial ecosystems can cause soil acidification, lead to eutrophication of sensitive waterbodies, affect biodiversity, lead to secondary N_2O emissions and cause changes in terrestrial carbon sinks (Liu et al., 2019; Elser et al., 2009; Zhan et al., 2017; Wang et al., 2017; Sutton et al., 2014). Liu et al. (2022) reported that global agricultural NH_3 emissions and N deposition increased by 78% and 72% from 1980 to 2018, respectively. Understanding the sources of NH_3 emissions and temporal and spatial patterns of N deposition will be essential for developing abatement strategies.

Reductions in NH_3 emissions from livestock via improved feed efficiency and enhancing NH_3–N capture from livestock manure and recycling that N back into cropping systems can help improve the overall N use efficiency and circularity of agricultural systems. In addition, development of enhanced efficiency bio-based fertilizers that are stable and release N slowly, as well as improved N management (better matching N to crop needs over time, immediate incorporation of fertilizers into soils, etc.), can also help minimize NH_3 volatilization losses in cropping systems.

Nitrate is one of the most widespread groundwater contaminants in the world, and its consumption has been linked with a variety of human health ailments. The main sources of NO_3 pollution of groundwater are from intensive use of N fertilizers (both synthetic and livestock manures) in agricultural production, disposal of sanitary and industrial wastes, leakage from septic systems and landfills and NO_x air stripping waste from air pollution control devices (Bhatnagar and Sillanpää, 2011; Abascal et al., 2022). Transport of NO_3 from ground waters to surface waters as well as runoff of NO_3 to surface waters has led to eutrophication in N limited systems, resulting in hypoxia and anoxia, loss of biodiversity and harmful algal blooms damaging fisheries and pristine marine environments (BijaySingh and Craswell, 2021). Optimizing N application rates and timing can be effective approaches to reducing NO_3 losses to ground and surface waters. In addition, development of more stable bio-based fertilizers with slow N release characteristics can reduce losses by better matching N release with plant uptake.

Nitrous oxide is a potent GHG as well as a stratospheric ozone depleting substance with a lifetime of 116 years (Prather et al., 2015). The largest global source of anthropogenic N_2O emissions is agriculture, representing 52% of total emissions (Tian et al., 2020). Tian et al. (2020) reported that the largest increases in anthropogenic N_2O emissions in the last four decades were due to direct emissions from agricultural soils receiving N additions (71%). There is also considerable positive feedback between climate change and increasing N_2O emissions. As the recent growth in N_2O emissions have exceeded some of the highest projected emission scenarios, mitigating these emissions is urgent (Gidden et al., 2019). Reducing excess N application to croplands and adopting precision fertilizer application methods provide the greatest immediate opportunities for the abatement of N_2O emissions.

Improving the connectivity between agricultural byproducts and agricultural production can improve N use efficiency and reduce N_r losses through more efficient recycling of nutrients. Application of bio-based fertilizers following the 4R strategy of applying the right fertilizer at the right rate and time with the right application method can enhance overall system efficiencies and thereby reduce their environmental impact.

2.1.2 Phosphorus losses

Globally, P deficiency is considered a major limitation for crop production, particularly in low-input agricultural systems (Raghothama, 2005). However, in many developed countries, over application of P as fertilizer (synthetic and bio-based fertilizers) has become a significant source for water pollution. In areas with concentrated livestock production, managing manure P to reduce negative environmental impacts is a challenge. Although manure P availability is greater or similar to synthetic fertilizer P use, manure P is not efficiently used in crop production. The inefficient use of manure P is due to several factors: uneven distribution of manure by grazing animals, incomplete collection and inadequate storage of manure from housed animals, poor timing of manure application, high cost of transporting manure, manure N:P ratios that do not necessarily match N:P needs of crops resulting in overapplication of P in some instances and the relatively low price of synthetic P fertilizer. Due to the high moisture content and bulky nature of manures, they are generally land applied within close proximity of where they are produced which had resulted in the buildup of P in soils surrounding livestock farms. This excessive P application has led to P surpluses in croplands, decreased P use efficiency and increasing P losses to surface waters.

Elevated P concentrations in receiving waters can lead to eutrophication, which can be costly to remediate. For example, in England and Wales, it has been estimated that damages due to agricultural losses of P are near £19 million (Bateman et al., 2011). In some countries, direct discharge of P in wastewater to surface waters is still common. For example, in Thailand, P containing wastewaters discharged directly to surface waters from dairy and swine farms were estimated to add 554 and 261 T P y^{-1} (Prathumchai et al., 2018). These discharges have a direct negative impact on surface water quality in these regions. Phosphorus surplus has been shown to increase with increasing livestock density in studies at continental, national and regional scale, especially at livestock densities above 2 LSU ha^{-1} (Liu et al., 2017; Nesme et al., 2015). One of the main causes of these surpluses is the large amount of P imported in feed coupled with low P use efficiency of most livestock. Therefore, there is often a clear relationship between livestock density and P balances at the farm level. In several countries (e.g. the United States, Netherlands, Norway, Denmark, Finland), manure P generated can meet or even exceed that needed for sustainable crop production in some regions (Smit et al., 2015; Hanserud et al., 2016; Yang et al., 2016; Parchomenko and Borsky, 2018; Svanbäck et al., 2019). Despite this large potential for within country P recycling, areas with the largest amounts of manure P are not necessarily co-located with the highest P needs. Therefore, hot spots occur where manure P exceeds that regionally needed for crop production. In addition, fertilizer P is still often used in these production systems leading to a further buildup of soil P.

Development of technologies to capture and concentrate P from agricultural byproduct streams will be essential to enable more sustainable recycling of P back through the production system. Additionally, cost-effective methods for redistribution of byproduct P from areas of surplus to areas of deficit will need to be developed. In some instances, particularly in livestock dense regions, regulations, economic incentives and technical solutions for enhanced relocation of byproduct P from areas with surplus to areas with deficit will be crucial. Support for processing and trading of byproduct-based P fertilizers can help reduce nutrient imbalances between areas with excess nutrient and those needing this valuable nutrient.

2.2 Heavy metals

Heavy metal contamination of byproducts used as bio-based fertilizers is a concern as these metals can bioaccumulate in agricultural crops and subsequently endanger the health of the consumer (Polechónska et al., 2018). Two of the most common byproducts used as bio-based fertilizers are manures and sewage sludges which may be applied raw or composted. On average, sludges have the highest values of heavy metal concentrations, compared to composts and manure, particularly when wastewater treatment plants also collect industrial effluents (Lopes et al., 2011). Cajamarca et al. (2019) characterized several agro-industrial, livestock and food wastes used to create bio-based fertilizers and found that some material contained high levels of trace metals, in particular cadmium (Cd). Livestock manures can contain elevated levels of arsenic (As), copper (Cu) and zinc (Zn) as these are found in feed additives to improve overall health and growth of livestock (Bloem et al., 2017). Repeated application of these bio-based fertilizers to cropland can lead to buildup of metals in soils which can potentially enter the food chain. Monte Carlo simulations indicated that repeated application of bio-based fertilizer likely increased the concentrations of Zn, Cd and As in soil compared with soil background levels (Gong et al., 2019). Concern over heavy metal contamination is ubiquitous where sludges, manures, composts and other bio-based fertilizers are applied to agricultural lands resulting in legislation in many countries regulating application (Lopes et al., 2011; Shi et al., 2018; Gong et al., 2019; Nunes et al., 2021).

In China, it was reported that livestock manure was one of the predominant sources of trace metals entering agricultural soils, accounting for approximately 55%, 69% and 51% of the total Cd, Cu and Zn inputs, respectively (Luo et al., 2009; Shi et al., 2018). Tan and Tran (2021) reported that Cu and Zn accumulated in soil, water and rice with application of sewage sludge at varying levels with concentrations exceeding permissible Vietnamese standards (QCVN 03: 2008) and US EPA 503 criteria. A risk assessment focused on reuse of organic waste as bio-based fertilizers, indicated that Zn was the main contributor to total risk due to its high concentration in bio-based fertilizers and high bio-transfer potential (Lopes et al., 2011). While others reported that health risk index values for Cd, cobalt (Co) and lead (Pb) suggested that these metals had the probability to cause health problems in people who consume vegetables grown with bio-based fertilizers (Ugulu et al., 2021). Others have reported heavy metal contents of bio-based fertilizers being below regulatory thresholds (Nekvapil et al., 2021; Golovko et al., 2022). In addition to health concerns, elevated levels of heavy metals in agricultural soils can result in decreased germination, altered metabolism, growth reduction, reduced biomass production and reduced yield for sensitive plants (Sethy and Ghosh, 2013; Goyal et al., 2020). While phytoremediation of metals from soils is feasible, there are few remediation strategies available, therefore prevention is key.

2.3 Pathogens

Pathogens can be present in food wastes, manures and other byproducts that are used as bio-based fertilizers. This contamination can pose potentially serious risks to human health as the application of these bio-based fertilizers to croplands can result in foodborne and waterborne outbreaks. The risks will vary from low to high, depending upon a number of factors, such as animal health, microbial concentration, land application method and environmental conditions (Venglovsky et al., 2009; Adegoke et al., 2016). Some pathogens commonly found in manures and food wastes are *Campylobacter*, *Escherichia coli* O157:H7, *Clostridium*, *Listeria*, *Salmonella*, hepatitis E virus, *Cryptosporidium parvum*, *Giardia lamblia* and *Shigella* (Kraus et al., 2003; Pepper et al., 2006; Manyi-Loh et al., 2016; O'Connor et al., 2022). The level of these zoonotic pathogens can exceed thousands per gram of material, with infection causing temporary illness or mortality, especially in high-risk individuals (Hutchison et al., 2005; Klein et al., 2010; Létourneau et al., 2010). Exposure of humans to pathogens can occur through occupational and recreational exposures, ingestion of contaminated food and water or aerogenic routes (Matthews, 2006; Dungan, 2010). According to the Centers for Disease Control and Prevention (CDC), *Escherichia coli* O157:H7, *Listeria monocytogenes* and *Salmonella* spp. are three major foodborne pathogens (CDC, 2020). These bacteria have been associated with numerous outbreaks in the United States (Ratnam et al., 1988; Nightingale et al., 2004; CDC, 2020).

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Application of contaminated bio-based fertilizers to soils, particularly surface application, can result in the transport of pathogens to surface or ground waters (Abu-Ashour et al., 1994; Jamieson et al., 2002; Tyrrel and Quinton, 2003; Bloem et al., 2017). The overland transport of microorganisms is called horizontal movement, while the leaching of microorganisms through soil and other porous subsurface strata is referred to as vertical movement. Unless a soil is saturated or contains an impermeable barrier, vertical movement of microorganisms will occur (Mawdsley et al., 1995). Some physical and chemical properties that influence the vertical movement of microorganisms are soil type, water content and water flow, microbe and soil particle surface properties, cell motility, pH, plant roots, temperature and presence of micro- and meso-faunal organisms (Mawdsley et al., 1995; Unc and Goss, 2004).

Rapid horizontal transport of microorganisms to surface waters can occur when either the rainfall intensity exceeds the soil's infiltration rate or when the soil becomes so saturated that no rainfall can percolate (Tyrrel and Quinton, 2003). Factors that influence the level of microbiological contamination in runoff from agricultural lands are organism die-off rates, quantity and type of amendment applied, sloping terrain, rainfall intensity and water infiltration rate (Evans and Owens, 1972; Doran and Linn, 1979; Baxter-Potter and Gilliland, 1988; Abu-Ashour and Lee, 2000; Jenkins et al., 2006; Ramos et al., 2006). Methods to mitigate the offsite transport of microorganisms in runoff from amended soils include use of vegetative filter strips (Coyne et al., 1995; Fajardo et al., 2001) or vegetative treatment systems with a settling basin for solids collection and a vegetated area (Koelsch et al., 2006; Berry et al., 2007).

Alternatively, bio-based fertilizers can be treated prior to land application, thus reducing subsequent risks associated with pathogens (Lund et al., 1996; Tiquia et al., 1998). Various physical, chemical and biological treatment technologies could be used to reduce or eliminate the presence of pathogens (Heinonen-Tanski et al., 2006). While there are advantages and disadvantages with these methods, some can provide additional benefits, such as the production of compost that can be used to enhance the properties of agricultural soils (Tester, 1990) or biogas for energy generation (Holm-Nielsen et al., 2009). There are a wide variety of technologies available to treat byproducts; however, the only processes with a documented record of cost-effective pathogen reduction are composting and anaerobic digestion (Sobsey et al., 2006; Martens and Böhm, 2009; Gurtler et al., 2018; Jiang et al., 2020; Thakalie and MacRae, 2021).

2.4 Antibiotics, antibiotic resistant bacteria and antibiotic resistance genes

In many countries, antibiotics are used therapeutically (high doses) in livestock production to treat specific diseases or subtherapeutically (low doses) by incorporating into feed to improve growth efficiency (Sarmah et al., 2006). It is also common practice to simultaneously administer multiple classes of antibiotics to livestock at the production facility (Song et al., 2007). Because not all antibiotics are absorbed in the gut of animals, they are excreted via urine and feces in unaltered form and as metabolites (Halling-Sørensen et al., 1998; Boxall et al., 2004). It has been estimated that as much as 80% of orally ingested antibiotics can be excreted in urine and feces (Elmund et al., 1971; Levy, 1992; Halling-Sørensen et al., 2002). Several classes of veterinary pharmaceuticals and antibiotics, including coccidiostats, ionophores, lincosamides, macrolides, sulfonamides and tetracyclines, have been detected in surface waters adjacent to livestock operations (Campagnolo et al., 2002; Hao et al., 2006; Song et al., 2007). In addition, the practice of land applying livestock manure as a bio-based fertilizer provides for the introduction of antibiotics over large areas in the environment, resulting in frequent detection of these compounds in soils and waters worldwide (Hamscher et al., 2002; Christian et al., 2003; Kemper, 2008).

Antibiotics can select for ARB in the animal gut, which are then released into the environment via excreted feces. ARGs are the genetic code ARB use to produce proteins responsible for antibiotic resistance. ARGs can be distributed to similar, distantly related and pathogenic bacteria through horizontal gene transfer mechanisms (Alekshun and Levy, 2007) and are considered an emerging contaminant (Pruden et al., 2006). Prophylactic and therapeutic uses of antibiotics in food–animal production has the potential to contribute to drug resistant bacteria in vivo, during manure storage, and in soils receiving manure solids or wastewater as a fertilizer (Binh et al., 2008; Negreanu et al., 2012; Chantziaras et al., 2014). Use of antimicrobials in livestock production may also intensify the resistance of pathogens to antibiotics, reducing the ability to treat infected individuals (Boxall et al., 2003; Bahe et al., 2006).

The continued use of large quantities of antibiotics in animal production and human health raises concerns about the release of these drugs and the increasing prevalence of ARB and ARGs in the environment when livestock manure and municipal biosolids are used as bio-based fertilizers. While use of antibiotic drugs can enrich ARB, the interplay between antibiotic drug use, ARB/ARGs and land use practices in agroecosystems is poorly understood (Williams-Nguyen et al., 2016). Manure application can transfer ARB and ARGs to soils, as well as antibiotic residues and other xenobiotic compounds, resulting in the expansion of antibiotic resistance reservoirs when compared to that of native soils (Heuer and Smalla, 2007; Cytryn, 2013; Amarakoon et al., 2016; Dungan et al., 2019). The detection of ARGs in soils, manures and agriculturally impacted environments is well documented in the scientific literature, but the risk of elevated ARG levels on public health is not well understood. Low concentrations of antibiotics and their metabolites can enter the food chain when plants are grown on fields which have received bio-based fertilizers contaminated by antibiotics (Bloem et al., 2017). This could potentially contribute to antibiotic resistance in humans and animals when edible crops are contaminated by traces of antibiotics, as it has been shown that even low-level antibiotic concentrations can select for antibiotic resistance in bacteria (Sandegren, 2014).

To minimize the effects of antibiotics, ARB and ARG in bio-based fertilizers, removal of these contaminants prior to land application may be necessary. Removal of antibiotics during anaerobic digestion has had varying success, even within the same class of antibiotics, while composting has been shown to significantly reduce antibiotics in nearly all cases (Youngquist et al., 2016). The removal of ARB and ARGs with digestion and composting varies with treatment effectiveness dependent on temperature (Beneragama, et al., 2013; Diehl and La Para, 2010; Guan et al., 2007; Resende et al., 2014). Alternatively, bioremediation using algae (Guo and Chen, 2015; Yu et al., 2017; Waseem et al., 2017) and fungal (Naghdi et al., 2018) species have successfully removed antimicrobials in water treatment facilities.

2.5 Chemicals of emerging concern

The trace levels of emerging or recently detected pollutants in soil and water receiving bio-based fertilizers is a growing concern for human health and the environment because at high concentrations, these chemicals can affect animal, human and environmental health. This category of emerging contaminants consists of pharmaceuticals, pesticides, hormones, disinfectants and their metabolites (Chaturvedi et al., 2021; Rathi et al., 2021). Hormones may be naturally occurring endogenous hormones, or natural or synthetic exogenous hormones used as growth promoters. Hormones found in animal waste are a concern as they may act as endocrine disruptors and influence the production of natural hormones, providing the blockage, minimization, stimulation or their inhibition (Matthiessen et al., 2002). Hormones have been found in high concentrations in pig farming effluent (Honorio et al., 2019; Fine et al., 2003). Hutchins et al. (2007) reported estrogens and estrogen metabolites in swine sow, finisher and nursery lagoons, as well as dairy, beef and poultry lagoons. Some hormones have even been detected in groundwater near livestock wastewater lagoons (Fine et al., 2003; Bartelt-Hunt et al., 2011).

Pesticides and microplastics are commonly found in food wastes due to their presence in soils and on foods (Nguyen et al., 2020; Tian et al., 2022). Some pesticides adsorb strongly to soil particles and can accumulate in soils and potentially be taken up by plants (Li et al., 2018). Pesticide residues that persist in bio-based fertilizers can damage soil quality as well as plant health

(Boudh and Singh, 2019). Composts containing residual herbicides were found to inhibit seedling vigor, emergence rate and plant growth in sensitive plants when mixed with soil at rates of 10–20% (Chang et al., 2017). Some plant growth regulator herbicides do not break down easily and can pass through animal digestive systems and composting facilities and have the potential to cause significant damage to plants and crops.

A wide range of microplastics composed of polyethylene, polypropylene, polyamide, polyester, polystyrene, polyethylene terephthalate, polyvinyl chloride, polyurethane, polyvinylidene chloride and polyacrylonitrile have been identified in the environment. Polymer-coated, slow-release fertilizers, plastic mulching for weed and pathogen control and other cultivation practices are important sources contributing to microplastics in soil. Composting, which usually reduces CEC in manures and agricultural wastes, accelerates microplastic formation when larger plastic contaminants become more fragmented. Although ingestion by earthworms and other soil fauna would be the most direct biological effect, microplastic's capacity to serve as particles capable of concentrating other contaminants, such as antibiotics and pharmaceutically active compounds, may be an unrecognized contributor to CEC risk.

PCBs are a widely recognized environmental and food contaminant, with human exposure primarily coming from consumption of animal-derived food such as meat, dairy and eggs, and fishery products (Weber et al., 2018). Animal feeds and feed additives are major sources of dioxins and PCB contamination for food of animal origin. Animals that are exposed to contaminated soil, such as beef cattle and veal from suckler cow herds, sheep and free-range chickens, have the highest risk of exposure through uptake of contaminated soil, grass, silage and hay. Application of sewage sludge and contaminated sediment deposits on agricultural land caused elevated PCB levels in meat from sheep and beef cattle grazing the contaminated areas (Weber et al., 2018). Management recommendations include not grazing contaminated areas, feeding non-contaminated feed sources during the finishing phase prior to slaughter, increasing the cutting height of plants grown on contaminated soil to reduce exposure to contaminated soil and mixing non-contaminated feed with contaminated feed sources to dilute the PCB in the whole feed. PAHs are classified as carcinogenic, mutagenic, teratogenic and immunotoxic to micro- and macro-organisms (Patel et al., 2020). PAHs are commonly found in aquatic and terrestrial environments and are strongly adsorbed to soil particles where they can enter the food chain due to plant uptake of contaminated soils (O'Connor et al., 2022).

Assuming increasing use of bio-based fertilizers for food production in the future, it would be beneficial to have concentration limits for CEC. Risk estimation based on risk quotients (RQs) indicated generally low environmental

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risks associated with application of bio-based fertilizer to soils for food crop production. However, the toxicity and potential concentration and build-up in soils of CEC mixtures needs to be considered when estimating the risks from application of bio-based fertilizers on agricultural land or in other production systems.

3 Case study: intensive dairy production in the northwest United States

3.1 Background

Intensive livestock production regions generate large volumes of manure that can be recycled back through crops, creating more sustainable circular systems. However, the large volume of manure and the potential contaminants found within can also create challenges. As an example, we will discuss several aspects of an intensive dairy production region and issues regarding sustainability/circularity of using manure as a bio-based fertilizer within the region. Idaho is the third largest dairy producing state in the United States with 70% of the lactating herd or ~894 000 animals (including mature and young stock) located in South Central Idaho, also known as the 'Magic Valley' region (Fig. 2). This region has approximately 429 000 ha of cropland resulting in an animal density of ~2 AU ha^{-1} which has been shown to be the stocking rate at which regional nutrient surpluses can occur (Liu et al., 2017; Nesme et al., 2015). All the manure generated within the region is land applied, with a majority applied either as a solid (some of which is composted) or a liquid (following solid separation) which is blended with irrigation water and applied through the irrigation system.

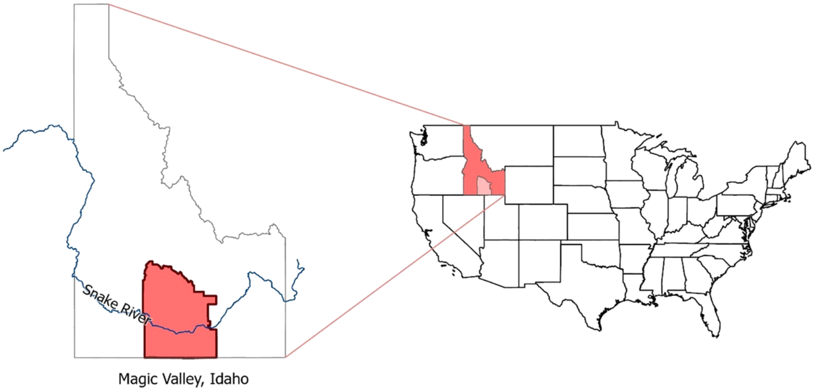

Figure 2 The Magic Valley region of South Central Idaho. This region is the third largest dairy producing region in the United States with ~894 000 animals (including mature and young stock) and approximately 429 000 ha of cropland resulting in an animal density of ~2 AU ha^{-1}.

This region has a semi-arid climate with an annual average precipitation of only 270 mm, which mainly occurs during the non-growing season. Therefore, all cropland in the region is irrigated with water diverted from the Snake River. This water is managed by the Twin Falls Canal Company (TFCC) south of the Snake River and the Northside Canal Company to the north. The TFCC routes irrigation water through 180 km of main canals and over 1600 km of smaller channels and laterals. Irrigation water flows by gravity from the Snake River throughout the 82 000 ha watershed (Fig. 3). Natural channels or coulees often convey water to laterals and collect runoff and return flow from fields. These return flows ultimately convey water back to the Snake River and can carry nutrients and other contaminants that may impair water quality. This irrigation tract has served as a benchmark watershed for the USDA Agricultural Research Service since 2004 and provides a unique system in which to evaluate the impact of land management practices on water quality in the region. In this section, we discuss the impacts of the recycling of manure products on nutrient balances and losses as well as the impact on antibiotic resistance in the regional environment.

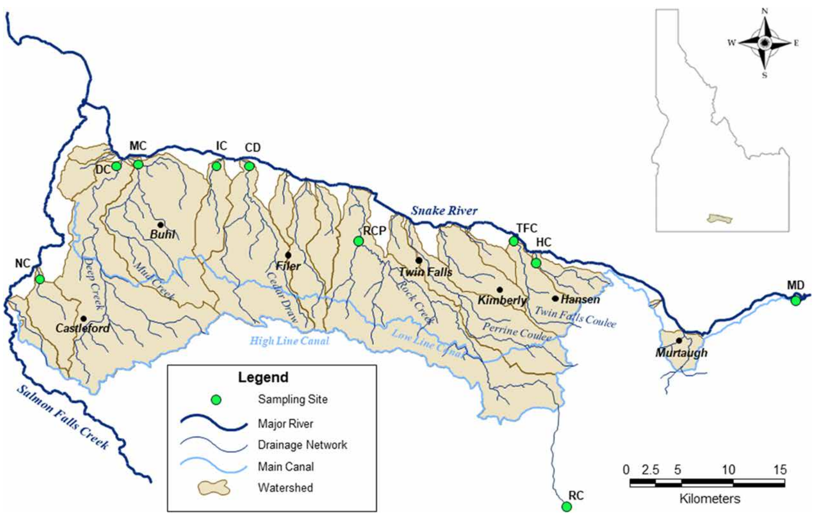

Figure 3 The Twin Falls Canal Company irrigation system including sampling sites for studies located within the watershed. The sampling sites within the Upper Snake Rock Watershed in South Central Idaho: N coulee (NC), Deep Creek (DC), Mud Creek (MC), I Coulee (IC), Cedar Draw (CD), Rock Creek Poleline (RCP), Twin Falls Coulee (TFC), Hansen Coulee (HC), Milner Dam (MD) and Rock Creek (RC). Both MD and RC were upstream sites and used to determine background concentrations. The NC and other return streams flow in a northerly direction into Salmon Falls Creek and Snake River, respectively. The Snake River flows to the west.

3.2 Impact of intensive dairy production on regional nutrient balances and losses of reactive nitrogen

Dairy production in the Magic Valley region of Idaho is characterized by high animal density at the regional (~2 AU ha^{-1}) as well as the farm level, where typical densities are closer to 4–20 AU ha^{-1}. Due to the import of nutrients via feed, on-farm surpluses of approximately 11 kg P cow^{-1} $year^{-1}$ and 174 kg N cow^{-1} $year^{-1}$ (including unaccounted for N) have been reported (Hristov et al., 2006; Spears et al., 2003). Unaccounted for N in these budgets are associated with losses of volatile N in housing and manure storage, which are ~50% of N excreted in some cases, resulting in N deficits for on-farm forage production (Leytem et al., 2018). Modeling of representative farms in the region has indicated that 70% of farms cannot meet crop N needs with manure only and need to import fertilizer N, while 80% of dairies produced more P on-farm than can be used by growing crops (Dell et al., 2022). These farm gate P surpluses have led to a buildup of soil P on many producer fields as transportation of manure longer distances is cost prohibitive. The Snake River, which receives runoff from agricultural fields in the region, has many segments of the river which have been identified as impaired with respect to P, resulting in total maximum daily loads (TMDLs) being set at 0.075 mg L^{-1} for total P (TP; IDEQ, 2010). In response, the Idaho State Department of Agriculture (ISDA) has required dairies in the state to develop a nutrient management plan to regulate, in particular, the amount of P being land-applied and evaluate the risk of potential P losses to both surface and groundwater via use of a P Site Index (Leytem et al., 2017).

Leytem et al. (2021) investigated the potential to recycle nutrients generated by dairy cattle in the region within the current agricultural system to better balance nutrients produced with regional crop demands, potentially ameliorating negative environmental impacts. They reported that manure N and P accounted for 45% and 55% of total regional inputs, respectively, with the balance consisting of fertilizer and biological N fixation (15%). There was a regional N surplus of 24 172 MT N $year^{-1}$, when accounting for all N inputs, compared to crop removal, and accounting for losses of both manure and fertilizer N as NH_3 (Fig. 4a). Overall, N losses as NH_3 were estimated to be approximately 44 000 MT N $year^{-1}$, representing 47% of N removed by crops each year. There was a regional P surplus of 8913 MT P $year^{-1}$ (Fig. 4b). However, when assessing P application per ha of cropland reported as receiving manure, the average surplus was 146 kg P ha^{-1} $year^{-1}$. For both N and P, there was a positive relationship between nutrient surpluses and dairy cattle populations ($r^2 > 0.9$). If manure N could be distributed across the regional cropland, then manure N could supply 44% of crop demand. However, it is important to keep in mind that 100% of the manure N is not available the year it is applied

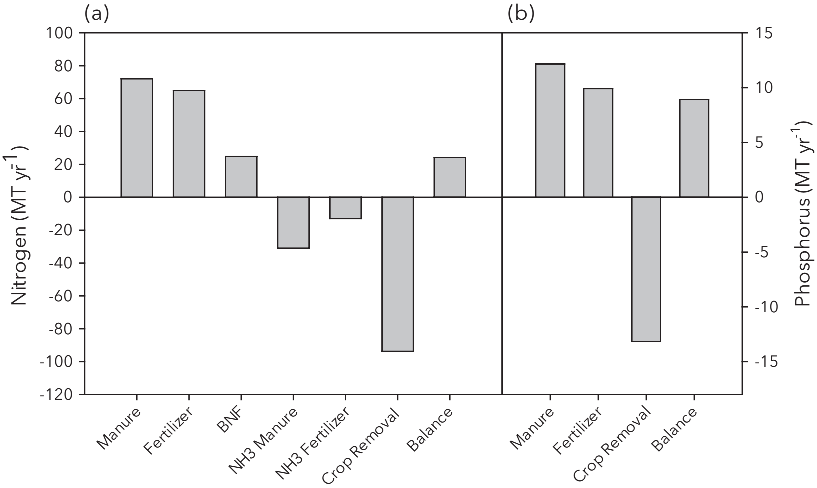

Figure 4 Regional nitrogen (a) and phosphorus (b) balances within the Magic Valley of South Central Idaho, an intensive dairy production region. Balances include N and P applied with manure and fertilizer and removed through crop uptake. Biological nitrogen fixation (BNF) was included as a source of N and losses of N as, ammonia (NH_3), from manure and fertilizer were also included in the balance.

and can vary substantially due to manure type, soils and climate. Average N mineralization rates for this region for solid manure (which represents roughly 67% of total manure) range between 15% and 30% (Leytem, unpublished data) each year. However, over time this N may mineralize and become available for plant uptake or losses to the environment. If N lost as NH_3 could be captured and re-used, then manure N could supply 77% of crop uptake. If manure P were distributed across the regional cropland, it would account for 92% of crop uptake and could replace synthetic fertilizer on most of the acreage. Utilizing the available road system, manure would need to be transported approximately 12.9 km to efficiently cycle it through the regional cropping system. However, due to fields that already have excess P and are not suitable for additional P applications, this transportation distance is likely greater. At present, the average maximum manure transport distance is 6–7 km from the dairy, therefore cost-effective methods for redistribution of manure P from areas of surplus to areas of deficit would need to be developed.

Losses of reactive N in this region have the potential to negatively impact the environment. Regionally, 44 000 MT N year^{-1} is lost as NH_3–N from agricultural production and there is potential for redeposition of N in areas where it could have negative impacts on N sensitive ecosystems. The Magic Valley is surrounded by native rangeland and is upwind of Yellowstone and Grand Teton National Parks, both of which are sensitive to N inputs. Estimating transport and deposition of this NH_3–N has been a new area of investigation in the region.

Utilizing a regional NH_3 monitoring network, estimates of N fluxes indicated a deposition rate of 10 to 40 kg N ha^{-1} $year^{-1}$ in areas impacted by agriculture and dairy production (Leytem et al., unpublished) while the surrounding natural environment may be sensitive to inputs as low as 2 kg ha^{-1} $year^{-1}$. These additions of N can lead to water quality issues and ecosystem impacts in downwind regions. With a regional N surplus of 24 172 MT N $year^{-1}$, water quality is also a concern. The USGS evaluated nitrate trends in groundwater over the periods 2000 to 2020 and found increasing trends in regions dominated by intensive agriculture/dairy production, although a majority of wells tested have NO_3 + NO_2 concentrations less than 10 mg L^{-1}, which is below the EPA threshold for drinking water (Skinner, 2022). Lentz et al. (2018) reported that NO_3–N in shallow groundwater in the region increased 1.4-fold from the late 1960s to early 2000s. Even though most well NO_3 concentrations are below the current U.S. EPA threshold, there is concern about increasing trends over time and what that might mean for groundwater quality in the future.

In the Magic Valley region, given the large annual inputs of N, there is potential of significant N_2O losses which can have negative impacts on climate change. To evaluate these potential losses, plot scale research has been conducted within the region to determine the effects of manure/fertilizer application, crop rotation and climate on N_2O losses. In general, fluxes of N_2O-N were greatest when both soil moisture and temperature were high (0.35 m^3 m^{-3} and 25°C), with a few large emission pulses accounting for the majority of growing season N_2O losses (Dungan et al., 2017, 2021; Leytem et al., 2019). In general, the addition of stacked solid manure had greater N_2O emissions than fertilizers or composted manures (Dungan et al., 2017). This enhanced loss of N_2O carried over beyond the year of application with higher losses from treatments receiving manure application 2 years previously. Growing season N_2O emissions also increased significantly with increasing manure application rate (Leytem et al., 2019). The amount of N_2O–N lost as a percentage of the total N applied across multiple studies ranged from <0.01% to 0.41% (Table 1). Regionally, losses of N_2O from application of manure and fertilizer would contribute approximately 1179 MT N_2O–N $year^{-1}$ (assuming 1% loss) to the atmosphere contributing to climate change.

Despite the higher N_2O emissions from manure application, soil organic carbon (SOC) was found to have increased in the soil profile (0–91 cm), while fertilizer treatments had a slight net loss of SOC (Bierer et al., 2021). This increase in SOC can help offset GHG emissions; however, SOC levels begin to decrease once manure applications are terminated, thus the offset will certainly decrease with time since manure was last applied. In addition to providing a potential GHG offset, studies have shown that manure can improve soil health by slowing down or reversing declining organic matter levels. The effect of manure application on soil health was evaluated in the top 30 cm of soil using 12 different biological

Table 1 Emission factors for nitrous oxide losses (N_2O) within the magic valley of southern Idaho

Reference	Treatment	%N_2O–N of TN applied
Dungan et al., 2017	Urea[a]	0.21
	SuperU[b]	0.09
	Compost[c] (34 Mg ha^{-1} annually)	0.09
	Fall manure[d] (52 Mg ha^{-1} annually)	0.09
	Spring manure (52 Mg ha^{-1} annually)	0.12
Leytem et al., 2019	Low manure application rate (18 Mg ha^{-1} annually)	0.13
	Low manure application rate (36 Mg ha^{-1} biennially)	0.15
	High manure application rate (52 Mg ha^{-1} annually)	0.18
	Urea	0.24
Dungan et al., 2021	Urea	<0.01
	Super U	<0.01
	Compost (34 Mg ha^{-1} annually)	0.06
	Fall manure (52 Mg ha^{-1} annually)	0.13
	Spring manure (52 Mg ha^{-1} annually)	0.41

Emission factors are presented as the loss of N_2O-N as a percentage of the total amount of N applied and represent the average (or cumulative) losses over multiple years.
[a] Urea fertilizer only.
[b] SuperU [(stabilized granular urea with urease {*N*-[*n*-butyl]-thiophosphoric triamide) and nitrification (dicyandiamide) inhibitors}].
[c] Composted dairy manure scraped from open lots and composted in windrows.
[d] Solid manure scraped from open lot dairies applied in the fall (after harvest) or in the spring (prior to planting) with target application rates of 18, 36 and 52 Mg ha^{-1} annually (dry weight basis).

and chemical indicators (Dungan et al., 2022). These metrics are commonly used to quantify organic matter pools and biological nutrient cycling via enzyme activities and N transformation rates. Compared to synthetic fertilizer, application of manure had a significant positive effect on most indicators, which increased with increasing manure application rate. While these results suggest that manured soil is healthier than soil receiving synthetic fertilizer, the caveat is that applying high rates of manure for many years has resulted in the gradual increase of N and P in the top 30 cm compared to synthetic fertilizer. As previously discussed, this buildup of N and P can pose a threat to water quality.

3.3 Impact of intensive agricultural production on determinants of antibiotic resistance in the environment

While land application of manure can improve soil fertility and health, there are growing concerns over the impact of this practice on the development and dissemination of antibiotic resistance in the environment (Fig. 5). An

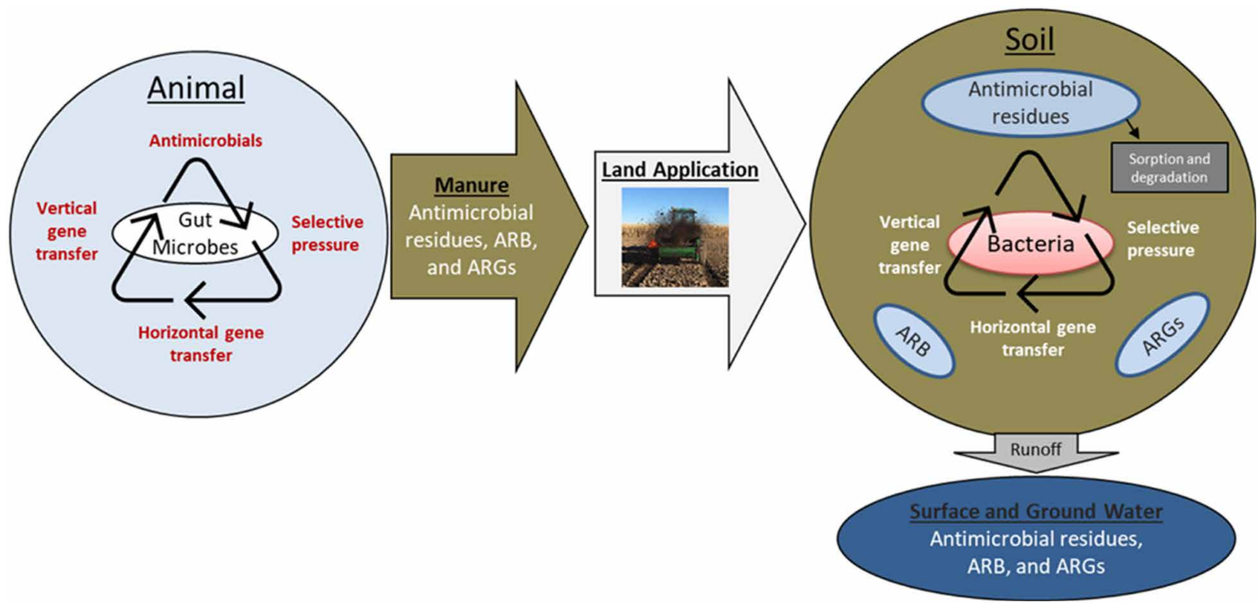

Figure 5 A conceptual diagram showing the potential fate of antimicrobial residues and mechanisms by which antibiotic resistance can be acquired by bacteria within the animal gut and in soil following the land application animal manure. Animal manure application is a major pathway by which antimicrobials, antibiotic resistance genes and bacteria are introduced into the soil environment. Figure adapted from Chee-Sanford et al. (2009).

investigation into the effect of solid dairy manure application on the abundance of ARGs during crop production was performed at plot scale (McKinney et al., 2018). Some ARGs detected at higher relative abundances (normalized to the 16S rRNA gene) in soil with manure application were those encoding resistance to sulfonamide and tetracycline antibiotics. In general, it was found that (1) manure application significantly increased ARG abundance in the top 30 cm of soil compared to plots receiving synthetic fertilizer only, (2) gene abundance increased with increasing manure application rate and (3) subsequent annual applications of manure did not increase the gene abundance above that of the first application.

Dairy wastewater is also commonly applied to agricultural soils; thus, it represents another mechanism by which antibiotic resistance determinants can be disseminated in the environment. To improve knowledge on this topic, a small-scale plot study was established to determine the effect of straight or diluted dairy wastewater on the abundance of ARGs (Dungan et al., 2018). The relative abundance of most ARGs increased dramatically after wastewater irrigation, compared to irrigation of control plots with canal water, and high gene levels were maintained throughout the 6-month study period. The results from this study suggest that increased ARG abundance was by addition of intracellular and extracellular genes present in the wastewater, not by enrichment of ARB. However, it was not known if the presence of antibiotic residues and other xenobiotic compounds may have affected gene selection and persistence in this study. What was clear from these results was that wastewater irrigation dramatically increased ARG abundance in soils receiving straight or diluted wastewater. The increase of the ARG reservoir is a potential cause for concern, as it could facilitate acquired resistance in bacteria that are pathogenic to humans and food-producing animals.

Moving beyond plot-scale studies, research was conducted to determine the abundance of several ARGs in agricultural and non-agricultural soils under various land use practices in the region, including cropland, forestland, inactive cropland, pastureland, rangeland, recreational and residential lands (Fig. 6, Dungan et al., 2019). Soil samples were obtained from 96 sites within seven counties in South Central Idaho. ARGs were detected in many of the soils (15 to 58 out of 96 samples), with a sulfonamide resistance gene being detected the most frequently (60% of samples). All the genes were detected more frequently in the cropland soils (46 sites) and at statistically greater relative abundances than in soils from the other land use categories. When the cropland gene data were separated by sites that had received dairy manure, dairy wastewater and/or biosolids (27 sites), it was revealed that the genes were found at statistically greater abundances (7- to 22-fold higher on average) than in soils that were not treated. This study provided evidence that agricultural production, especially use of manure and biosolids, is contributing to the

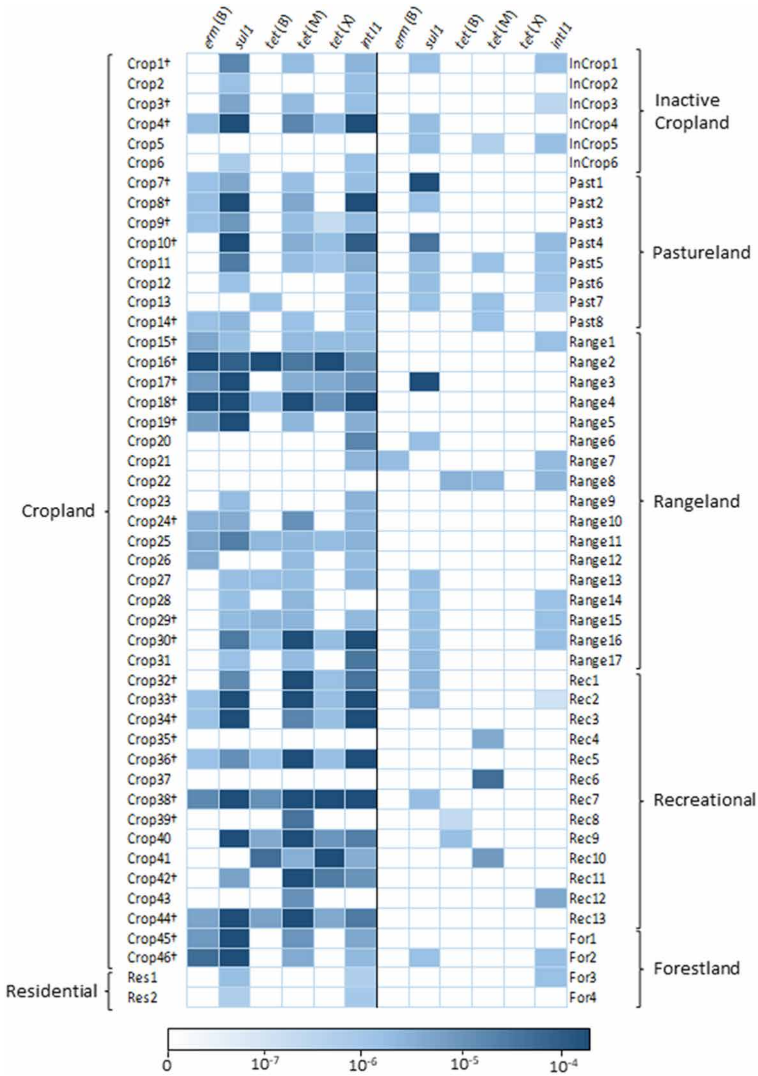

Figure 6 A heat map illustrating the relative abundance of selected antibiotic resistance genes in soils by land use category. The tetracycline resistance genes cover three known mechanisms of resistance including efflux [*tet*(B)], ribosomal protection [*tet*(M)] and enzymatic [*tet*(X)]; *sul1* represents a main determinant of sulfonamide resistance in Gram-negative bacteria; *erm*(B) confers resistance to macrolide-lincosamide-streptogramin B (MLSB) antibiotics and *intI1* is a class 1 integron-integrase gene and is linked to antibiotic resistance genes. The color scale at the bottom indicates the gene abundance from no detection (white) to ≥10^{-4} gene copies/16S rRNA gene copies (dark blue). †Indicates cropland soils that have a history of receiving dairy manure, dairy wastewater and/or biosolids.

expansion of antibiotic resistance-related determinants in regional cropland soils.

Land application of manures and biosolids containing antibiotic residues can pose a risk for transport of these residues to surface water via runoff (Joy et al., 2013; Soni et al., 2015; Bartelt-Hunt et al., 2009; Dolliver and Gupta, 2008; Kay et al., 2004). Antibiotics associated with clinical uses can also reach the environment from municipal wastewater treatment plant effluent discharges (Batt et al., 2006; Michael et al., 2013). Regardless of source, antibiotic contamination is recognized as being widespread in water resources. The spatial and temporal occurrence of 21 antibiotics in eight irrigation return flows (IRFs) within the TFCC irrigation tract was investigated (Dungan et al., 2017). Seven antibiotics of veterinary and human origins were detected at frequencies ranging from 3.1% to 62.5%. Monensin, which is exclusively used in cattle and poultry production, was the most frequently detected antibiotic and it was present in all IRFs (Fig. 7). This result suggests that monensin, and other veterinary antibiotics, are entering return flows in runoff from fields that had received livestock manure or wastewater. Some antibiotics were also detected at the site of irrigation water diversion from the Snake River. Therefore, even cropped soils that are not treated with manure, wastewater or biosolids are still receiving low-level antibiotics during irrigation events. The occurrence of antibiotics at low concentrations may pose a health risk to humans and animals due to the proliferation of antibiotic resistance in the environment (Kemper, 2008).

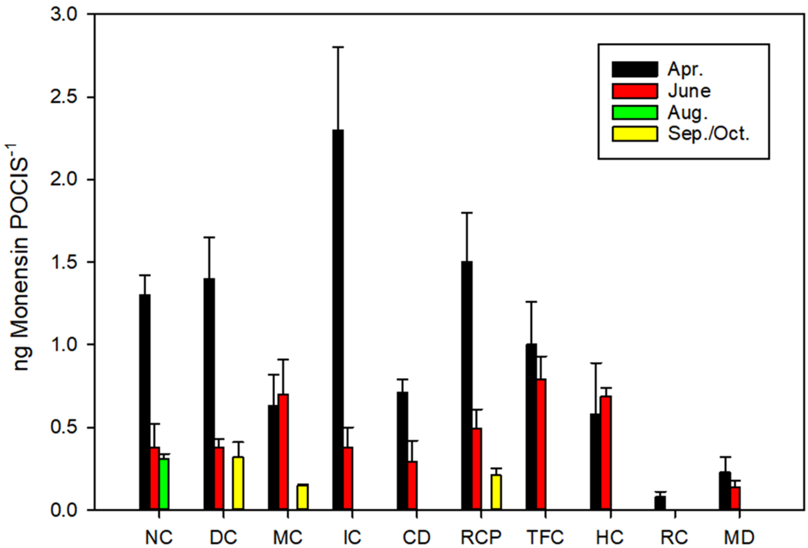

Figure 7 Mass of monensin extracted from polar organic compound integrative samplers (POCIS) that were deployed at the sampling sites within the Upper Snake Rock watershed in South Central Idaho. Columns represent means ± SD (n = 3).

Surface waters can be a dominant route by which ARGs and ARB are disseminated in the regional watershed. A study was conducted to determine the abundance of selected ARGs in the IRFs that transport irrigation water from agricultural fields back to the Snake River (Dungan and Bjorneberg, 2020). The ARGs were recovered at all IRF sampling sites with detections ranging from 55 to 81 out of 81 water sampling events. When compared to the average annual relative gene abundances in the canal water samples, they were found to be at statistically greater levels. The fact that most IRFs contained higher levels than found in the canal water indicates that IRFs can be a source of ARGs that ultimately discharge into the Snake River.

The antimicrobial susceptibility among *E. coli* and enterococcal isolates that were obtained from the regional watershed were determined (Dungan and Bjorneberg, 2021). *E. coli* and enterococci are important indicators for understanding the impact of fecal pollution on water resources but reports on antimicrobial resistance among these organisms in IRFs are lacking. Environmental isolates can be a potentially important source of AMR and return flows may be one way resistance genes are transported out of agroecosystems. For *E. coli*, 75% of the isolates were pan-susceptible, while 13% of isolates were resistant to tetracycline and fewer numbers being resistant to 13 other antimicrobials. For the enterococcal species, only 9% of isolates were pan-susceptible and the single highest resistance was to lincomycin (75%). Furthermore, 17 *E. coli* and 13 enterococcal isolates were found to be multidrug resistant to up to seven different drug classes. These results indicate that the IRFs are polluted with material of fecal origin, which would not be surprising given that this is a mixed-use watershed and livestock manures are commonly applied to cropland soils. However, some of the *E. coli* and enterococcal isolates could be from naturalized extraintestinal populations that were released from sediments and soils. Regardless of fecal bacteria source, a wide variety of resistance patterns were found among many of the isolates, suggesting the potential for horizontal transfer of ARGs in the aquatic environment.

4 Conclusion

The valuable nutrients and C found in many agricultural byproducts have the potential to be recycled as bio-based fertilizers within the greater agricultural system, thereby enhancing the circularity and sustainability of this sector. While the potential for beneficial use of these products is great, there are potential risks associated with their use that must be carefully considered. Balancing nutrients in areas with intensive livestock production, avoiding health risks associated with bioaccumulation and biomagnification of heavy metals as well as reducing exposure to pathogens, antibiotics and other CEC are imperative for these products to be used safely and sustainably. Careful risk assessment and management/treatment may overcome most barriers for reuse of these valuable products enabling greater use throughout the agricultural sector.

5 Future trends in research

To further improve the recycling and reuse of agricultural byproducts as bio-based fertilizer in food production systems, further research is needed to address several areas of concern. Improved nutrient use efficiency and reduction of losses, in particular N and P, to the environment are greatly needed. Technologies that allow capture and stabilization of N and P from livestock manure over a range of farm sizes are needed to more effectively recycle these nutrients back to cropland that is deficient in these nutrients. This also includes methods to condense nutrients to reduce the cost of transporting them from regions of surplus to those of deficit. Improved models for estimating crop N uptake to better match N additions are necessary to enhance overall N use efficiencies. Improved models are also needed to estimate N and P losses to the environment to better predict the effects of management strategies on air and water quality as well as losses of N_2O that may impact climate change.

In livestock systems, alternatives to the use of metals (Cu and Zn) and antibiotics in diets as growth promoters are necessary to reduce their potential negative impacts when manure is used as a fertilizer in the landscape. Effective treatment technologies are needed to remove metals, kill pathogens, remove or inactivate antibiotics, and other CECs from agricultural byproducts that may be used as bio-based fertilizers are needed to ensure that these products can be safely and sustainably used within the food production system. Improved risk assessments to establish critical levels of these potential contaminants are also needed to provide guidance for reuse of byproducts as bio-based fertilizers.

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