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ENVIRONMENTAL ISSUES

The emergence, trajectory, and impacts of emerging contaminants publications in the *Journal of Environmental Quality*

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Abstract

As analytical capabilities in the early 2000s began to enable the detection of chemicals in environmental media at increasingly small concentrations, chemicals with the potential to cause adverse human and ecosystem health effects began to be found nearly ubiquitously worldwide. The types of chemicals that were targeted for analysis included natural and synthetic hormones, human and veterinary pharmaceuticals, chemicals in personal care products, novel pesticides, nanoparticles, microplastics, and other chemicals of natural and synthetic origin. The impacts of these chemicals on environmental and human health in many cases remain unknown. Collectively, these chemicals became known as "emerging contaminants" or "contaminants of emerging concern." Much progress has been made toward understanding the sources of these contaminants in the environment, the processes that control their fate and transport once they are released into the environment, and the ability of technology and/or best management practices to mitigate their occurrence. As the *Journal of Environmental Quality* (JEQ) celebrates its 50th anniversary, we sought to understand how publications in the journal have made impactful contributions in the research area of emerging contaminants. Here, we present the trajectory of publications in JEQ that have shaped knowledge in this field, highlight the importance of these contributions, and conclude with opportunities for JEQ to continue attracting high-quality emerging contaminants research.

1 | INTRODUCTION

Contaminants of emerging concern, or emerging contaminants, are defined as natural and synthetic compounds that can potentially have harmful impacts on human and ecosystem health, typically at trace concentrations, and that are not currently regulated by environmental and water quality laws. These compounds have been detected throughout the environment, especially in waterbodies and soil, in urban and agricultural areas. These chemicals originate from human and animal waste, including industrial waste and application of pesticides. Existing treatment processes of domestic and industrial wastewater are often unable to remove these emerging contaminants prior to their discharge into natural waters and application onto land.

Abbreviations: BMP, best management practice; PFAS, per- and polyfluorinated alkyl substances; PPCPs, pharmaceuticals and personal care products.

The beneficial reuse of human and animal wastes has long been viewed as an environmentally responsible practice that reduces reliance on unsustainable depletion of freshwater resources and synthetic fertilizers. However, the land application of human and animal residuals inadvertently introduces other chemicals into the environment, particularly agroecosystems, that are known or suspected endocrine disruptors or retain potency. These chemicals are then suspectable to mobilization during rainfall events, causing them to leach to underlying groundwater aquifers and/or be transported via surface runoff to nearby surface water bodies. Repeated application over long periods of time, as is common in agricultural fields and in some urban areas, has the potential to lead to accumulation of these emerging contaminants, thereby reducing the ability of impacted soils to act as effective biogeochemical filters. While the fate and transport of nutrients, sediment, and conventional pesticides in the environment are generally well known and have been published in the *Journal of Environmental Quality* (JEQ) since the journal began, the processes driving the fate and transport of these emerging contaminants only began to be studied in the late 1990s and early 2000s.

The nationwide reconnaissance study conducted by the USGS in 1999–2000 (Koplin et al., 2002) on the occurrence of emerging contaminants downstream of wastewater treatment plants and agricultural activities is generally considered to be the seminal paper that initiated an exponential growth of papers published on emerging contaminants in aquatic ecosystems. With more than 5,300 citations and nearly 34,000 article views, it maintains its place as the most highly cited paper in *Environmental Science & Technology* nearly 20 years after it was first published (Westerhoff et al., 2020). However, the first JEQ paper in the research area that would eventually become known as emerging contaminants was published a few years earlier. Tanghe et al. (1999) were interested in understanding the sources of potential toxicity of estrogenic activity in surface water and sewage treatment plants. This trio of Belgian researchers sought to identify the sources of estrogens and estrogen-mimicking compounds that were being found in aquatic ecosystems and causing alarm among toxicologists. Their research found that industrial wastewater contained nonylphenol, a nonionic surfactant used in industrial processes, detergents, and cleaning products and categorized as a "xeno-estrogenic compound." Tanghe et al. (1999) found that the industrial wastewater released into the aquatic ecosystem contained enough estrogenic activity to cause concerns for aquatic organisms. Their sampling of waters in agricultural land uses and downstream of domestic wastewater treatment plants did not appear to have elevated concentrations of this chemical, and therefore the concern was primarily linked to industrial sources.

This early research on the sources and potential impacts of endocrine disrupting compounds was striking, particularly because water samples were quantified for a target analyte of concern but also because assays were used to determine estrogenic activity in the sample, despite not necessarily knowing which chemicals contributed to the estrogenicity. This type of approach would soon become widespread in emerging contaminants literature, as researchers began to make links between contaminant sources, environmentally relevant concentrations, and threats that the presence of these contaminants posed to sensitive, nontarget aquatic organisms. Further, hormone-focused research has continued, with now more than two decades of research focused on sources, impacts, and solutions to the pervasive issues of elevated natural and synthetic hormones in the terrestrial and aquatic ecosystems.

Core Ideas

- This paper examines the history and impact of emerging contaminants papers published in JEQ.
- 67 papers published by JEQ are identified as within the umbrella of emerging contaminants.
- Major categories of papers included hormones, PPCPs, nanoparticles, and novel pesticides.
- The top 10 most highly cited papers are featured and their impacts on the field discussed.
- Future opportunities for JEQ to continue to impact this research area are presented.

Between 1991 and June 2021, JEQ published 67 papers related to emerging contaminants (Figures 1 and 2). This includes 39 papers focused on pharmaceuticals and personal care products (including hormones and antibiotics); 14 papers focused on nanoparticles; 7 papers focused on topics ranging from endocrine disrupting compounds, volatile organic compounds, and greenhouse gases; 6 papers focused on pesticides; and most recently, 1 paper focused on the emerging area of microplastics.

Here, we present an overview of the emerging contaminants papers that have been published in JEQ over the past two decades, trace the impact these papers have made on the field, and highlight the important ways that JEQ has improved knowledge on the sources, fate, transport, and impacts of emerging contaminants in the environment while remaining true to its core mission of understanding anthropogenic impacts on the environment, especially in the context of agroecosystems.

2 | HORMONES

True to the traditional types of nutrient-centric papers published by JEQ, the research on emerging contaminants starting in the early and mid-2000s focused on the introduction of

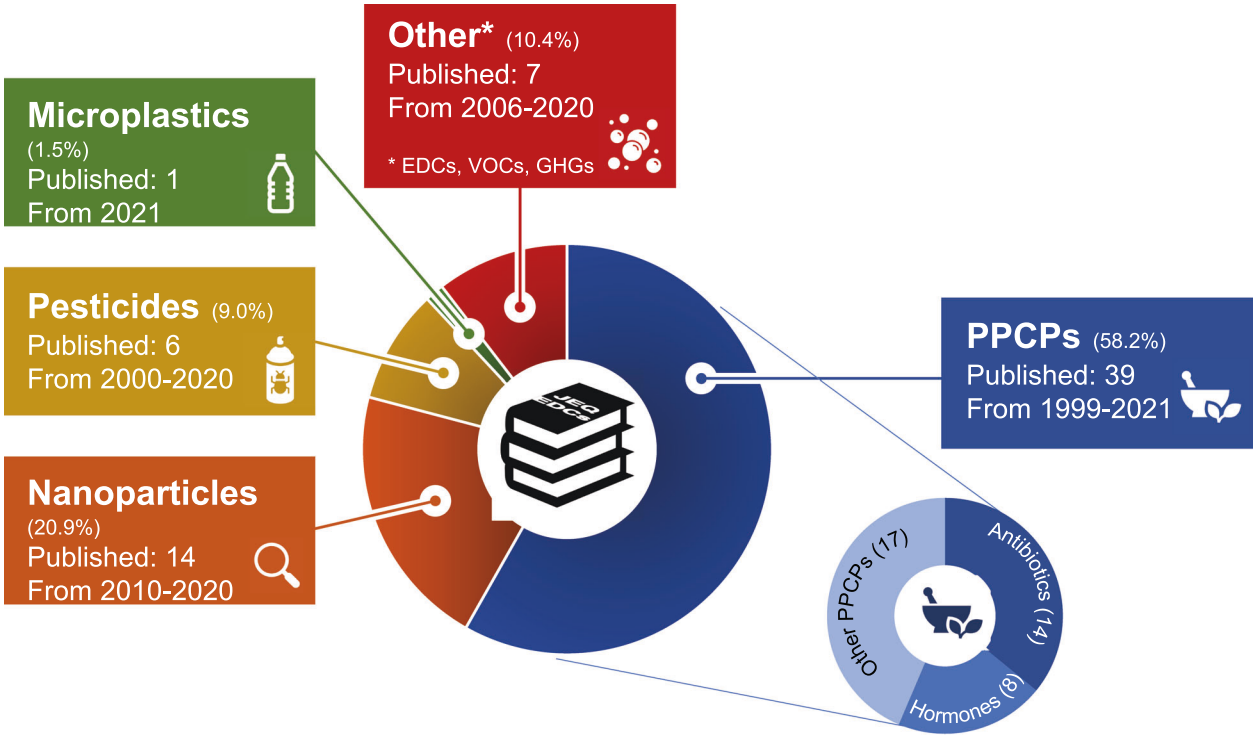

FIGURE 1 Manuscripts published in the *Journal of Environmental Quality* from 1999 to June 2021 focused on emerging contaminants and grouped by major topic. The percentage of each topic represents the percentage of publications within each topic area relative to the total number of papers published on emerging contaminants ($n = 67$) in this time period. EDCs, endocrine disrupting compounds; GHGs, greenhouse gases; PPCPs, pharmaceuticals and personal care products; VOCs, volatile organic compounds

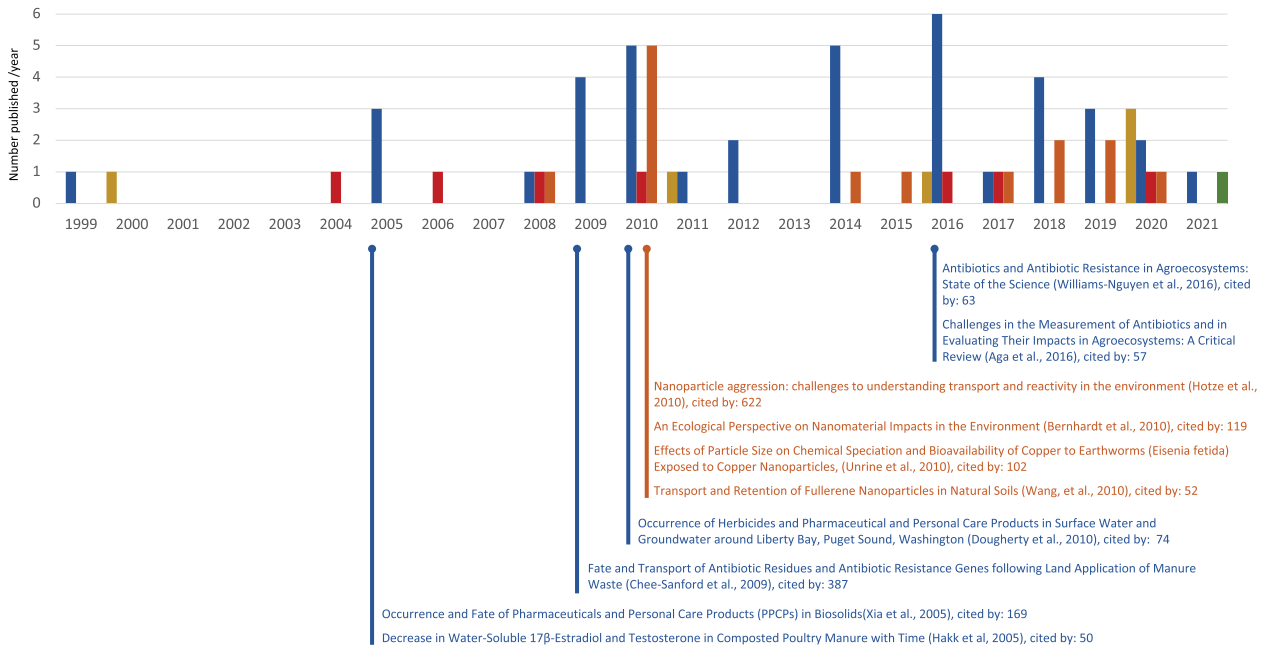

FIGURE 2 Timeline of papers published in *Journal of Environmental Quality* (JEQ) from 1999 to 2021 focused on emerging contaminants. Each bar represents the number of papers within the five topic areas: pharmaceuticals and personal care products, pesticides, nanoparticles, microplastics, and other, in a given year. The call-outs represent the top 10 cited papers

natural chemicals as a result of the land application of wastewater beneficial reuse (i.e., wastewater spray irrigation) and manure and best management practices (BMPs) that are effective in reducing the environmental impact of these contaminants. Research on the presence and transport of hormones present in reclaimed water and manure have had a relatively long publication record in JEQ, with papers spanning from 2005 to the present. Research published in JEQ found several effective strategies to reduce the transport of manure-borne hormones from agricultural fields, including composting poultry manure prior to land application (Hakk et al., 2005) (Figure 2), pelletizing poultry litter prior to application (Dutta et al., 2010), and land-applying dairy manure via subsurface injection rather than surface broadcasting (Mina et al., 2016). Of course, livestock are not the only sources of hormones in the environment, and Woodward et al. (2014) demonstrated that long-term spray irrigation of treated wastewater can be a source of natural and synthetic hormones into agroecosystems, as wastewater treatment plants are unable to effectively remove hormones from influent wastewater. However, their work also suggested that agricultural soils can act as effective biogeochemical filters, reducing the hormone concentrations within the first meter of the soil profile. Agricultural soils were also found to reduce estrogenic activity in treated wastewater by more than 200% by Singer and Brown (2018). These papers all actively sought ways to demonstrate that despite the potential threats that hormones pose to ecosystem health, there are effective ways to continue to reuse these beneficial resources as nutrient sources in environmentally responsible manners.

3 | PHARMACEUTICALS AND PERSONAL CARE PRODUCTS

The presence of human and veterinary pharmaceuticals in wastewater, biosolids, and manure have also continued to raise concerns about their potential impacts on organisms that these chemicals were never designed to affect (i.e., nontarget organisms). Concerns, especially on nonlethal behavioral changes that these chemicals can induce, sparked research published by JEQ starting in 2002 and continuing through the present. Given these new concerns, some of the earliest research published by JEQ focused on improving our knowledge on the occurrence of pharmaceuticals and personal care products (PPCPs) in human and animal residuals. In 2005, Xia et al. (2005) and Guerra et al. (2015) quantified the occurrence of trace organics in biosolids and biosolids production in wastewater treatment plants, respectively. The findings published by Xia et al. (2005) were highly impactful on the field, gathering widespread attention and more than 150 citations. Importantly, citation records reveal that while the Xia et al. (2005) paper was highly cited (Figure 2), papers that cited it were even more highly cited, with some, such as Santos et al. (2010) and Watkinson et al. (2007) each receiving >1,000 citations.

A few years later, Chee-Sanford et al. (2009) (Figure 2) linked the presence of trace-level pharmaceuticals in manure to the presence of antibiotic-resistant bacteria and genes also being introduced into agricultural fields, posing risks to microbial biodiversity and other aspects of ecosystem and human health. This publication was cited more than 380 times, and those papers were also cited more than 10,000 times collectively. For example, papers that cite Xia et al. (2005), such as Davies and Davies (2010), Laxminarayan et al. (2013), and Zhang et al. (2015), have received >4,000, >3,000, and >1,000 citations, respectively. It is clear that JEQ pushed the state of science forward significantly in this topic area, demonstrating the importance of understanding the fate and transport of emerging contaminants in agroecosystems to the broader goal of understanding emerging contaminants in the aquatic ecosystem.

Beyond characterizing the extent to which land-application of wastes introduced PPCPs into the environment, JEQ advanced knowledge on the fate of these chemicals once they were in the environment. In 2012, Williams and McLain (2012) examined the persistence of pharmaceuticals at a wastewater reuse site in arid Arizona and found that some pharmaceuticals accumulate near the soil surface, while others tend to be vulnerable to leaching, posing a threat to underlying groundwater aquifers that the wastewater was recharging. Other field studies also examined the potential for PPCPs to leach through soil following land-application, including on turfgrass fields (Gan et al., 2006; Bonderenko et al., 2012; Young et al., 2014). Franklin et al. (2016) demonstrated that pharmaceutical compounds that accumulate in the soil profile as a result of wastewater irrigation activities can also be taken up by crops, thereby introducing them into the food chain.

The potential for septic tanks to be sources of PPCPs and affect underlying groundwater has also been explored by papers published in JEQ (Katz et al., 2010; Kibuye et al., 2019; Spoelstra et al., 2017). These studies demonstrated the inability of regulations established primarily to protect groundwater from nitrate contamination to provide effective mitigation for some persistent or frequently used pharmaceutical compounds, and therefore PPCPs were found at elevated concentrations in potable groundwater supplies. These findings are of particular importance to rural households, which often rely on private wells as a drinking water supply.

While field-based studies provide important information about where and at what concentrations PPCPs are found in the environment, laboratory-based and modeling studies often provide clarity on the mechanisms and processes governing PPCPs transport that can be difficult to study in isolation in field experiments with less control and more variables. Laboratory-based studies conducted by Xu et al. (2009), Kwon et al. (2010), and Dalkmann et al. (2014a, 2014b)

characterized sorption, transport, and transformation of various PPCPs, providing a better understanding of the role of organic carbon, pH, aerobic and anaerobic conditions, and the physicochemical characteristics of the pharmaceuticals themselves on the fate and transport of PPCPs and their metabolites once they are land-applied to soils. These column and microcosm studies could then provide the information necessary to improve existing models, such as MACRO (Larsbo et al., 2009) and HYDRUS (Filipović et al., 2020) to better represent the transport of emerging contaminants.

Beyond basic science research on characterizing fate and transport dynamics, JEQ has also significantly advanced applied science in the topic of emerging contaminants. Questions regarding how land-application of PPCP-containing materials can be improved to reduce the environmental impact have been a prominent research focus of more recent papers in JEQ. Ray et al. (2017), Le et al. (2018), and Dolliver and Gupta (2008) assessed manure storage, management, and application methods that can reduce the losses of veterinary pharmaceuticals from agricultural fields, helping farmers to understand how strategies commonly used for reducing nutrient losses may also provide co-benefits for emerging contaminant loss.

4 | NANOPARTICLES

In the 2000s, research on nanotechnology, nanomaterials, and nanoparticles was growing exponentially (Bernhardt et al., 2010). At the time, there was optimism around the potential for these technologies and materials to significantly improve quality of life. For example, the antimicrobial properties of nanosilver made it appealing for inhibiting bacterial growth in food packaging and in the textile industry. However, presence of these nanoparticles in the environment poses concerns for ecological health and microbial biodiversity in the environment. One of the most impactful papers published in JEQ was a review article written by Bernhardt et al. (2010), in which these benefits and concerns were presented through an ecological framework that garnered more than 100 citations since its publication a decade ago. One of the papers citing Bernhardt et al. (2010), Yin et al. (2011), has received >500 citations. The theme is the same as PPCPs: products that were designed to make our lives better were causing problems in places that they were never intended to be found—and at such trace-level concentrations that only the most recently developed analytical tools and technology allowed us to detect them in environmental matrices.

In the most highly cited article on emerging contaminants published in JEQ, Hotze et al. (2010) reviewed the mechanisms governing nanoparticle aggregation and the complexities that these mechanisms caused for studies seeking to understand nanoparticle fate and transport in the environment. Beyond challenges posed by the chemical properties were those posed by the shape of nanoparticles, with some more spherical in shape and others shaped more like rods or tubes. This review paper of the current state of knowledge has more than 620 citations, well above any other emerging contaminants themed papers in JEQ. More recent citations of this article include those on the topic of micro- and nanoplastics, which face similar challenges as those posed by the nanoparticles Hotze et al. (2010) so thoroughly described, setting the stage effectively for future research not only on metal-coated nanoparticles but also on plastics. The success of this paper published in JEQ demonstrates the importance of basic science articles published by JEQ, in addition to the strengths JEQ has in advancing applied science.

Following the Hotze et al. (2010) and Bernhardt et al. (2010) articles, traditional laboratory-based studies were also conducted to try and fill the knowledge gaps on the processes governing nanoparticle fate and transport in the environment; however, these subsequent papers had less of an impact on the field than the 2010 papers, with most of the nanoparticle laboratory- and field-based papers in JEQ each receiving less than 40 citations. Interestingly, the laboratory-based studies generally received more citations than the field-based studies, which contrasts with other emerging contaminants papers published in JEQ. Transport of nanoparticles through columns were conducted to better understand the factors affecting mobility through different soils (Saleeb et al., 2019; Sekine et al., 2017; Wang et al., 2010; Zhang et al., 2018) and the time scales over which different physiochemical processes affected fate (Sekine et al., 2017).

Fewer field-based studies on occurrence and fate of nanoparticles were published in JEQ and were only recently published, thereby limiting the number of citations they have received to date. However, the main findings of these two field studies have the potential to be paradigm-shifting. River and Richardson (2019) found that phosphorus-containing nanoparticles comprised approximately half of the dissolved reactive P entering the western portion of Lake Erie, such that the fraction of P categorized as dissolved reactive P actually contains particulate-P, with the particulate-P at the nanoscale. Most recently, Taylor et al. (2020) demonstrated that long-term biosolids application at agronomic rates did not lead to exceedance of silver ecotoxicity limits, suggesting that current application recommendations were sufficient to protect agricultural fields from the potential adverse impacts of nanoparticles in the biosolids.

5 | NOVEL PESTICIDES

Beyond hormones, PPCPs, and nanoparticles, JEQ papers in the area of emerging contaminants have also begun to include novel pesticides. In 2016–2021, JEQ published four papers

on neonicotinoid insecticides, one of which was a laboratory-based bioassay study to understand how changing soil moisture conditions expected in future climate scenarios affect the toxicity of neonicotinoids to nontarget insects (Hennig et al., 2020). Beringer et al. (2021) determined partition coefficients (K_d and K_{OC}) for clothianidin in wetland soils to determine how oxidation–reduction conditions and organic carbon content of the soils affected clothianidin sorption. Frame et al. (2021) found subsurface flow pathways to be significantly more important to the overall transport of neonicotinoids leaving agricultural fields due to the pesticide being introduced into the agroecosystem below the soil surface as a seed coating, rather than applied at the surface.

Few papers in JEQ have addressed the fate, transport, and effect of novel pesticides in environmental matrices. However, the publication of three papers in 2020–2021 on neonicotinoids suggests that this may change in the future. This appears likely to be only the beginning of papers published in JEQ on neonicotinoids but suggests that similar field and laboratory-based approaches are likely to be published by JEQ in the near future, furthering our knowledge on the fate, transport, and inadvertent impacts of neonicotinoids in agroecosystems, similar to the types of papers published by JEQ on other emerging contaminants.

6 | FUTURE OPPORTUNITIES

As research on emerging contaminants continues, it appears clear that there are significant opportunities for JEQ to continue to lead in the knowledge regarding how novel contaminants are introduced into the environment, particularly in agroecosystems through beneficial reuse of human and animal waste residuals, the fate and transport of these contaminants once they have entered the environment, and the ecological and human health risks that these contaminants pose, especially in the aquatic environment. The most significant opportunities for JEQ are on the topics of microplastics and per- and polyfluorinated alkyl substances (PFAS). Wastewater and biosolids are known to contain these contaminants, and both pose potentially significant challenges to long-term beneficial reuse programs. Concerns regarding the presence of PFAS in rural water supplies recently led to the USEPA investing several millions of dollars into research on this topic in a Request for Applications it released in 2019, highlighting the need to understand how PFAS may adversely impact vulnerable rural populations. As of June 2021, no PFAS papers have been published in JEQ, and only one examining microplastics fate and transport has been published (Kuoppamäki et al., 2021).

Further, the impacts of the COVID-19 pandemic on the presence of PPCPs in wastewater influent and effluent are also potentially significant. Widespread and increased usage of sanitizers and cleaning products, along with increased usage of pharmaceuticals to treat patients during the pandemic likely has resulted in increased presence of PPCPs in wastewater destined for beneficial reuse. Additionally, novel and experimental treatments for COVID-19 patients could lead to the presence of pharmaceuticals such as remdesivir in wastewater and biosolids, establishing a need to understand the fate, transport, and impacts of novel pharmaceuticals in the environment.

Finally, the co-benefits of agricultural BMPs adopted for mitigating nutrients and sediment from agricultural activities is another emerging area of interest. Riparian buffers, constructed wetlands, and other structural BMPs likely provide mitigation of novel pesticides, hormones, and other emerging contaminants present in manure, biosolids, and treated wastewater. While JEQ has demonstrated the potential for manure management strategies typically adopted to reduce nutrient transport to also reduce antibiotic and hormone transport, research on the co-benefits (and potential adverse effects) of widespread adoption of new BMPs will advance understanding of the extent to which watershed management plans will also address concerns regarding emerging contaminants in the environment. Two decades of research on emerging contaminants published by JEQ show that while some basic questions have been answered about these contaminants using laboratory, field, and modeling studies, many questions remain. Answers to these questions, combined with current knowledge, are necessary to improve ecological and human health. Given that emerging contaminants are frequently introduced into the environment by application of wastewaters and residuals to agricultural soils, JEQ is a logical publication outlet for dissemination of these future research results.

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AUTHOR CONTRIBUTIONS

Heather E. Preisendanz: Conceptualization, Formal analysis, Supervision, Writing-original draft. Ryan G. Barnes: Data curation; Investigation; Visualization. Michael L. Mashtare: Conceptualization, Formal analysis, Supervision, Visualization, Writing-review & editing. Anna Lintern: Conceptualization; Investigation; Writing-review & editing. Odette Mina:

Formal analysis; Investigation; Supervision; Writing-review & editing. Clinton Williams: Formal analysis; Investigation; Supervision; Writing-review & editing. Herschel A. Elliott: Conceptualization, Supervision, Writing-review & editing.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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