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Detection, Occurrence and Fate of Emerging Contaminants in Agricultural Environments (2019)

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ABSTRACT: A review of 82 papers published in 2018 is presented. The topics ranged from detailed descriptions of analytical methods, to fate and occurrence studies, to ecological effects and sampling techniques for a wide variety of emerging contaminants likely to occur in agricultural environments. New methods and studies on veterinary pharmaceuticals, microplastics, and engineered nanomaterials in agricultural environments continue to expand our knowledge base on the occurrence and potential impacts of these compounds. This review is divided into the following sections: Introduction, Analytical Methods, Fate and Occurrence, Pharmaceutical Metabolites, Anthelmintics, Microplastics, and Engineered Nanomaterials.

Practitioner Points:

- New research describes innovative new techniques for emerging contaminant detection in agricultural settings
- Newer classes of contaminants include human and veterinary pharmaceuticals
- Research in microplastics and nanomaterials show that these also occur in agricultural environments and will likely be topics of future work

KEYWORDS: pharmaceuticals, steroid hormones, anthelmintics, nanoparticles, analytical methods, antibiotics, water and wastewater, agricultural environments.

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INTRODUCTION

Water resources in agricultural environments are impacted by a wide variety of contaminants including nutrients, sediments, and pesticides. These groups of contaminants typically occur at easily measured concentrations in surface run–off in agricultural watersheds.

Nutrients, especially nitrogen, and pesticides have also been shown to impact ground water quality in areas susceptible to contamination. The impacts of newer contaminant classes such as pharmaceuticals, antibiotics and nanoparticles are less well–known. These "emerging" contaminants clearly have potential to enter the environment and cause known or suspected adverse ecological or human health effects. Release of these contaminants to the environment has very likely occurred for quite some time, but methods for their detection at environmentally–relevant concentrations have only recently become available.

Evaluating the environmental fate and effects of emerging contaminants includes compounds such as surfactants, antibiotics and other pharmaceuticals, steroid hormones and other endocrine–disrupting compounds (EDCs), fire retardants, sunscreens, disinfection byproducts, new pesticides and pesticide metabolites, and naturally–occurring algal toxins. Detection of these contaminants in environmental matrices (water, wastewater, soils and sediments) is particularly challenging because of the low detection limits required, the complex nature of the samples, and difficulty in separating these compounds from interferences. New extraction and clean–up techniques, coupled with improvements in instrumental technologies provide the needed sensitivity and specificity for accurate measurement.

The objective of this paper is to review the literature published in 2018 evaluating the detection, fate, and occurrence of emerging contaminants, with a focus on those contaminants likely to be found in agricultural systems. Relevant contaminants are personal care products, antibiotics and other pharmaceuticals associated with wastewater, and microparticles. Studies on pesticides and flame retardants are not reviewed unless they were evaluated in the same study. A new section this year is a review of the literature on microplastics in agricultural environments.

ANALYTICAL METHODS

New developments in analytical methods permit more rapid, sensitive, and simplified analysis of emerging contaminants in agricultural environments. Refinement of extraction, purification and detection methods can support research in the fate, transport and biological effects of these substances.

Althakafy et al. (2018) described a method to determine personal care products and pharmaceuticals in freshwater invertebrates with high pressure liquid chromatography high resolution mass spectrometry (HPLC/HR-MS) detection in full scan mode. Detection limits for each analyte were < 2.38 ng/g while mass accuracy was mostly < 2 ppm. A QuEChERS^{TM} extraction method was optimized to provide acceptable recoveries of the target analytes from the organisms. Tandem mass spectrometry was utilized for confirmation using optimized collision energies. Analytes detected in the invertebrates were metformin, caffeine, and triclosan.

A semi-quantitative screening method for the determination of pesticides and pharmaceuticals in water was developed by Arsand et al. (2018). The method used solid-phase extraction (SPE) with LC interfaced to a quadrupole time of flight (qTOF) mass spectrometry to detect 170 analytes in wastewater and 198 analytes in surface water. Detection limits for the screening method

were generally below 1 µg/L for most target compounds with several compounds detected at the 0.01 µg/L level. Analysis of real samples revealed the possibility of detecting other untargeted analytes in the screening process using the capabilities of the qTOF high resolution mass spectrometer.

Bhalsod et al. (2018) studied the uptake of pharmaceuticals in lettuce by developing a modified QuEChERS^{TM} method to extract the target analytes from soil and the root and shoot of the plant. Irrigation water samples were extracted using HLB cartridges. HPLC with tandem quadrupole mass spectrometry (HPLC-MS/MS) was used to quantify the analytes after cleanup. Recoveries from the lettuce ranged from 72% to 96%. Overhead irrigation was determined to increase concentrations for trimethoprim, monensin sodium, and tylosin in the lettuce shoots but not in roots. Carbamazepine was found to be persistent in soil and accumulated in the lettuce at concentrations higher than other target analytes.

Castrignano et al. (2018) examined chiral and achiral fluoroquinolones in wastewater and solids using chiral HPLC-MS/MS to separate and quantify several of the individual enantiomers. Recoveries of > 70% were obtained for most analytes with detection limits < 7 ng/L for wastewater and < 4.9 ng/g for solids. The method was successfully utilized to determine the enrichment of *S*-(-)-ofloxacin and its metabolite's *S*-enantiomers in wastewater when compared to upstream waters where racemic (±)-ofloxacin was observed.

Pressurized accelerated solvent extraction (ASE) was utilized by Ezzariai et al. (2018) to determine macrolide, tetracycline, and fluoroquinolone concentrations in compost. Analytes were detected with ultrahigh pressure (UPLC) coupled to electrospray mass spectrometry but exhibited matrix effects, which the authors addressed by using standard addition to quantify the target compounds. Recoveries ranged from 25-30% for roxithromycin to 100-171% for ciprofloxacin. Results from the analyses indicated that the composting process was effective in removing macrolides and tetracyclines from the compost but had little effect on fluoroquinolone.

Kazakova et al. (2018) developed an optimized HPLC-MS/MS method with an enzymatic microwave-assisted liquid extraction to quantify fluoroquinolones, tetracyclines, sulphonamides, penicillins, non-steroidal anti-inflammatory drugs, and trimethoprim in crayfish (*P. clarkii*). The mass spectrometer was operated in both positive and negative modes for electrospray ionization. Detection limits of 0.6-12 ng/g with recoveries >70% were obtained. Addition of the enzyme (Proteinase-K) during extraction of the biological matrix increased recoveries by approximately 10% for each analyte. The method was applied to crayfish obtained from the Doñana Nature Reserve in Spain and two pharmaceuticals (sulfamethoxazole and flumequine) were detected.

Solid-phase extraction and UPLC-MS/MS detection are the basis of a multi-residue method described by Klančar et al. (2018). The method was validated for 44 pharmaceuticals with detection limits below 5 ng/L and extraction recoveries above 90%. Labeled internal standards were used to minimize matrix effects. Sixty-six percent of all target analytes were detected in every water sample

collected from six rivers and lakes in Slovenia with valsartan observed at concentrations up to 47 ng/L.

Kong et al. (2018) presented a screening method utilizing UPLC/MS/MS for pharmaceutical and other residues in carp, shrimp, crab, eel, and mussel. Extraction of homogenized organisms with EDTA-Na_2, acetonitrile, and ethyl acetate gave recoveries between 50% and 120% at 50 ng/g for 160 target analytes for carp samples and 176 analytes for shrimp. Stronger matrix effects contributed to varying recoveries for crab, eel, and mussel samples. At concentrations near 1 ng/g, the number of detectable target analytes is limited to between 45 and 100, depending on matrix. The method was applied to aquatic food samples in China where ethoxyquin, enrofloxacin, ciprofloxacin, and trimethoprim were observed most frequently.

Ciprofloxacin-molecularly imprinted polymers were developed by Li et al. (2018) to selectively sample for ciprofloxacin residues in water. Uptake of ciprofloxacin onto the polymer was linear over time and recovery was 96.8%. Comparisons of binding parameters with other drug residues for both imprinted and non-imprinted polymers are presented. Ciprofloxacin concentrations obtained from polymer and grab samples are shown to be in close agreement. Effects on the absorption of ciprofloxacin onto the polymer due to other environmental parameters such as pH and ionic strength were also studied.

Polyethersulfone microextraction of emerging contaminants was investigated by Mijangos et al. (2018a) to determine 41 pollutants in samples from seawater, wastewater effluents, and estuaries. LC-MS/MS detection with electrospray ionization was utilized for quantitation. The method was optimized for pH, ionic strength, EDTA quantity, and amount of fiber polymer. Recoveries of 80-119% were obtained with detection limits ranging from 0.4 ng/L to 26 ng/L which compared favorably to results from solid-phase extraction for each water matrix. The method was applied to Spanish water samples obtained from the Basque Country where effluent and estuary water concentrations up to 1096 ng/L were obtained for irbesartan with valsartan, acesulfame, and sucralose also observed at elevated concentrations.

Tang et al. (2018) explored the use of eco-friendly and hydrophobic deep eutectic solvents (DES) based on fatty acids and alcohols to extract levofloxacin and ciprofloxacin in water by liquid-liquid microextraction. Sixteen types of DES media with different configurations of hydrogen-bonding donors and acceptors were compared in efficiency of extraction and stability in water. The most stable and efficient DES was observed to be composed of a 1-octanol/tricaprylylmethylammonium chloride mixture. Optimization of extraction parameters resulted in extraction recoveries exceeding 94.8% for both target analytes.

Concentrations of sulfamethazine, tylosin and atrazine in a tile drained watershed were the focus of a study by Washington et al. (2018). The authors utilized polar organic chemical integrative samples (POCIS) to collect target analytes in tile drain effluent and surface water originating in animal feeding locations. POCIS sampling collected target analytes over a longer time frame than by grab sampling. LC-MS/MS was used to quantify the resulting extracts to obtain concentration values. Comparisons between the two collection methods revealed

that the POCIS samples had higher detection frequencies and lower estimated concentrations. Time weighted average concentrations of 1.87 ng/L, 0.30 ng/L, and 754.2 ng/L were observed for sulfamethazine, tylosin, and atrazine, respectively.

Wu et al. (2018) developed fluorescent ciprofloxacin-molecularly imprinted nanoparticles to measure ciprofloxacin concentrations in water. The method was linear up to a concentration of 250 nM with a detection limit of 4.04 nM using a fluorescence spectrophotometer. The method was applied to distilled water, tap water, and aquaculture water with excellent recovery. The procedure to synthesize the nanoparticles as well as thermal field emission SEM images and characteristic data from FTIR and x-ray photo spectroscopy of the nanoparticles are presented.

OCCURRENCE AND FATE OF PHARMACEUTICALS

The availability and persistence of six veterinary antibiotics were studied by laboratory incubation assays under various soil water holding capacities by Albero et al. (2018). Antibiotic concentrations were positively correlated with the dissolved organic carbon. Increased absorption of the antibiotics was observed in presence of composted manure amendment over the incubation time. Half-lives ranged from 8-27 days with the exception of fluoroquinolone, where 70% of the residues remained after 90 days. Sulfamethoxazole was found at the highest levels in the soil aqueous phase, followed by sulfamethazine and lincomycin, with total of 98.2% of all the antibiotics. Chlortetracycline, doxycycline, ciprofloxacin and enrofloxacin were ≤1.8%. The authors concluded that the route of entry of the antibiotics through recycled water or manure impact their availability in the soil systems.

Twelve commonly used antibiotics were detected by Azanu et al. (2018) from different wastewater sources and lettuce samples in farms in Ghana. Sulfamethoxazole, erythromycin, ciprofloxacin, cefuroxime and trimethoprim were frequently detected. Ciprofloxacin was found at the highest concentration at 15 µg/L in the hospital wastewater while the river water impacted by wastewater discharges had concentrations of 3 µg/L. Irrigation water samples had lower levels of antibiotic concentrations, up to 0.2 µg/L. Edible lettuce tissues had total antibiotic concentrations, ranging from 12.0 – 104 and 11.0–41.4 ng/kg.

Air and precipitation samples were analyzed by Ferrey et al. (2018) within the Twin Cities Metropolitan Areas of Minnesota, U.S., for 126 pharmaceuticals, personal care products and other commercial chemicals. Bisphenol A, *N,N*-diethyl-meta-toluamide (DEET) and cocaine were most frequently detected with maximum concentrations of 3.8, 9.49 and 0.171 ng/L respectively in snow, 0.137, 0.37 and 0.033 ng/m^3 in air and 14.5 ng/L of DEET and 0.806 ng/L of cocaine in the rain samples. Antibiotics such as oloxacin were detected in rain and air, and ciprofloxacin, enrofloxacin and sulfamethoxazole were detected in rain with concentrations up to 10.3 ng/L. Other compounds detected in snow included iopamidol and naproxen at concentrations ranging from 3.74-228 ng/L. Caffeine was detected in air at 0.069 ng/m^3. Benzotiazole, its derivatives were detected between 1.5 -70 ng/L in rain and snow. Nonyphenol and derivatives were detected between 0.032-0.165 ng/m^3 in air.

Land spreading of biosolids generating PM10 particulates is a suggested input source for air. Wastewater is a source for precipitation samples. Chemical profiles in precipitation and air match with surface water exhibiting the atmospheric transport of these chemicals.

In another study of selected Minnesota lakes, antibiotics were detected in sediment cores by Kerrigan et al. (2018). The cores successfully captured historical trends of 10 antibiotics, primarily derived from wastewater effluents. These chemical groups include 4 compounds from sulfonamides, 3 fluoroquinolones,1 macrolide, trimethoprim and lincomycin with concentrations ranging from 1.2 - 66.1 ng/g.

Hossain et al. (2018) investigated the surface water samples from old Brahmaputra river in Bangladesh and detected 9 antibiotics and metronidazole in all samples (100% detection), with concentrations ranging from 0.05 – 13.51 ng/L followed by trimethoprim with second highest detection at 95% and highest concentrations of 17.20 ng/L. Other pharmaceuticals detected included sulfonamides, macrolides and carbamazepine with detection frequency between 60-85% and their concentrations were higher in fed aquaculture areas. Other input sources include hospitals, nursing homes, sewage wastewater and surface runoff.

Several studies were reported from Spain in 2018. Biel-Maeso et al. (2018a) measured a wide range of pharmaceutical compounds in the wastewater treatment plants in south west Spain, to test the suitability of reclaimed wastewater for irrigation. The total concentrations in the influents were 73-372 µg/L and effluents 3-41 µg/L with similar chemical composition and hydrochlorothiazide and diclofenac being predominant. Soil samples had 2-15 ng/g of pharmaceuticals, predominantly analgesics and anti-inflammatories, followed by antibiotics and psychiatric drugs. Pharmaceuticals were detected to a depth of 150 cm, indicating their high leaching capabilities due to heavy precipitation. Winter months had higher concentrations of up to 14 ng/g.

In another study, Biel-Maeso et al. (2018b) evaluated 78 pharmaceuticals in different aquatic marine environments from the Gulf of Cadiz in south west Spain, and found higher pharmaceuticals in seawater ranging from 61-2133 ng/L compared to coastal and oceanic transects ranging from 16-189 ng/L. Risk assessment studies found no environmental risk in coastal and oceanic sampling areas. Among the pharmaceuticals, analgesics, anti-inflammatories followed by antibiotics were detected more frequently. Caffeine was also detected in the oceanic seawater.

Boy-Roura et al. (2018) conducted a field study on occurrence and fate of 53 antibiotics from 10 chemical groups in an alluvial aquifer originating from the manure application in north east Catalonia region of Spain. Eleven antibiotics from the chemical groups of fluoroquinolones, macrolides, quinolones and sulfonamides were detected in groundwater compared to 2 groups, fluoroquinolones and sulfonamides in surface water with highest concentrations of 300 ng/L. Sulfamethoxazole and ciprofloxacin had the highest detection frequency. Spatial trends were not evident and the occurrence was assumed to be based on physio-chemical properties of the antibiotics that impacted the sorption, desorption and degradation patterns.

In Basque Country, Spain, Mijangos et al. (2018b) evaluated wastewater treatment plant effluents adjacent to 3 estuaries. Anti-inflammatory drugs, hypertensive drugs and caffeine were detected in higher frequency using passive sampling techniques (POCIS). Risk assessment studies indicated, acute toxicity for the angiotensin blocker compounds. Chronic toxicity was estimated for caffeine, diclofenac, bezafibrate and sulfadiazine.

Several studies were conducted in various parts of China. Hong et al. (2018) detected 27 pharmaceutical compounds including tetracyclines, sulfonamides, macrolides, chloramphenicols and analgesics and anti-inflammatory drugs in the Jiulong River watershed in the southeastern part of China. Concentrations ranged from 450 ng/L to 19,810 ng/L and comparable to other studies from this region. Urban land use was associated with higher occurrence and concentration of the pharmaceuticals. Macrolides showed medium to high risk to aquatic species

Chen et al. (2018) studied the Hai River system in northern China, for occurrence and risk quotient of antibiotics. Sixteen antibiotics including sulfamethoxazole, norfloxacin, erythromycin and roxithromycin had higher risks to aquatic organisms and generally higher than global waters, hence identified as priority contaminants for better regulation.

Chen et al. (2017) collected groundwater samples from monitoring wells in northern and southwestern China. Seventy four monitoring wells contained at least one antibiotic with ofloxacin, lincomycin and norfloxacin having highest detection frequency of 64 -70% and elevated concentrations between 441.9 ng/L – 1199.7 ng/L. Shallow groundwater in southern China seemed to contain the most antibiotics due to high input concentrations in the discharge and frequent exchange of groundwater with surface matrices.

Jia et al. (2018) surveyed the urban river of Xi'an and found the effluents from the wastewater treatment plants contribute to the antibiotic pollution. 14 antibiotics from 7 chemical groups were detected with sulfonamides, quinolones, macrolides and tetracyclines at high frequencies between 85-100% with concentrations in water samples up to 0.08 ng/L and sediments 0.002 µg/kg. Seasonal variations were found, with higher concentrations observed in March compared to July. Redundancy analyses were performed to detect environmental factors influencing the occurrence, and anthropogenic factors were the highest contributors to the pharmaceutical inputs.

Lai et al. (2018) studied occurrence and distribution of 49 antibiotics, 49 pharmaceuticals and 12 industrial and residential compounds in 14 aquaculture sites and neighboring aquatic areas in Taiwan. Ibuprofen, lincomycin, flumequine, caffeine, ifosfamide and cephalexin were detected in higher frequencies (>70%) with concentrations ranging from 172 – 788 ng/L. At a lesser detection frequency and concentrations, sulfamethoxazole, erythromycin and perfluoroalkyl substances were also detected suggesting cross contamination of the aquaculture from human activities.

Lesser et al. (2018) surveyed 218 organic compounds in springs groundwater and wastewaters in the Mezquital Valley system, Mexico. 23 pharmaceuticals were detected in the groundwater sources, with frequent detection

of sulfamethoxazole, N,N-diethyl-meta-toluamide, carbamazepine, and benzoylecgonine (primary cocaine metabolite) in groundwater, suggesting that pharmaceuticals are still able to reach the aquifers even though the soils act as a filtering system. Endocrine disruptors, volatile organics and semi-volatile organics were also detected along with the pharmaceuticals with concentrations for the various chemical groups ranging from 0.65 -71.59 µg/L

Reis-Santos et al. (2018) investigated 66 veterinary and pharmaceuticals in surface waters of Tejo estuary in Brazil with collection sites covering the entire estuary including main river, inflows, wastewater treatment outfalls and the traversing urban, agriculture, aquaculture and natural reserve areas. Pharmaceuticals were found in all the sampling sites with antibiotics, such as beta blockers, antihypertensive and anti-inflammatories more frequently detected with frequency >90% and concentrations ranging from 15 – 351 ng/L. These results provide a baseline database for regulatory information and future evaluations.

In one of the first studies, antibiotics discharged from pharmaceutical manufacturer was studied in Hanoi, Vietnam, Thai et al. (2018). The water samples collected from the outlets of the manufacturing plants as well as a hospital and an aquaculture farm in the region. 10 antibiotics, including ampicillin, cefuroxime, cefotaxime, clarithromycin, azithromycin, sulfamethoxazole, trimethoprim, ofloxacin, norfloxacin, and ciprofloxacin; were detected in the samples at different concentrations ranging from 41-252 µg/L. The effluents from the pharmaceutical plants had higher concentrations than the other sources suggesting wastewater from manufacturing plants as a major source of pollutants. Quinolone and sulfonamides had the highest detection frequency and concentrations.

PHARMACEUTICAL METABOLITES

Biosolids and treated wastewater are increasingly being applied to agricultural systems to meet the growing demand for alternative sources of irrigation and fertilizers whilst offering a practical solution for wastewater treatment by-product disposal. These practices can however inadvertently introduce emerging contaminants into agricultural systems. To date, research has predominantly focused on the presence of veterinary medicines or human use pharmaceuticals (prescribed) in biosolids and treated wastewater, however, recent research has also identified the presence of illicit drugs and their metabolites in sludge (<245 ng/g) which would present a route by which this class of contaminants can enter and persist in agricultural soils. Based on the quantification of pharmaceuticals in sludge originating from five wastewater treatment plants, Ivanova et al. (2018) predicted pharmaceutical mass loads in agriculture via land application of sludge. Assuming no further degradation of pharmaceuticals between sludge production and field application an annual load to soil in the Slovak Republic was predicted to range from 0.005 kg/year (clonazepam) and 120 kg/year (fexofenadine) with illicit drugs comprising 0.5% of the total load.

The fate of emerging contaminants in agricultural soils has been shown to be strongly influenced by the type of soil amendment, soil properties and chemical properties as documented in a number of studies published in 2018. Studies designed to evaluate the fate of land-applied

emerging contaminants have primarily focused on a select number of antibiotics (tetracyclines, sulfonamides, fluoroquinolones, and macrolides) and the antimicrobial, triclocarbon whilst concentrating on contaminated biosolids as the pathway by which these chemicals enter the soil environment.

Geng et al. (2018) observed that the fate of ^{14}C-labelled N-acetyl sulfamethoxazole residues in agricultural soils amended with sludge depended on the treatment processes the sludge had undergone prior to incorporation with the soil (e.g. limed, heat dried). More than 80% of the initial ^{14}C-activity was no longer extractable after 14 days incubation, except in the soil with limed sludge where 23% was $CaCl_2$ extractable. Although ^{14}C residues were more extractable in soil with limed sludge they exhibited less mineralization. Specifically, liming and drying the centrifuged sludge decreased ^{14}C mineralization in soils from 5.7-6.4% to 1.2–1.8%. As soils amended with limed sludge contained more ^{14}C residues at the end of the experiment (119 days) and had a larger bioavailable fraction this suggests an increased potential for uptake and leaching in agricultural systems in comparison to the other sludge treatments evaluated in this study.

Research has also demonstrated the accumulation of antibiotic residues in Chinese agricultural soils resulting from repeated biosolid amendment Yang et al. (2018a). Interestingly the antibiotic residual concentrations in soils followed a similar order to the biosolid loadings with fluoroquinolones > tetracyclines > macrolides > sulfonamides. However, tetracycline residues in the soils were up to 10 times lower than those of the fluoroquinolones, even though the concentrations of tetracyclines and fluoroquinolones in the biosolids were similar. With the exception of oxytetracycline and roxithromycin, predicted half-lives for the antibiotics were generally in excess of one year with a maximum calculated half-life of 3.69 years. Given that the average application rate of biosolids was reported to be twice per year, half-lives greater than 1 year demonstrate the potential for antibiotics to persist in agricultural systems and increase in concentration over time. However, antibiotic dissipation rate was affected by the presence of metals (slowed dissipation rate) and suspension of sludge application (increased dissipation rate) which affects the bulk soil concentration of antibiotics.

Lozano et al. (2018) also observed that the period since the last biosolid application to soils occurred strongly influenced soil concentrations of the antimicrobial agent triclocarban based on soil samples collected from 26 commercial farms in the US. In fact, triclocarban was not removed in soils where the time since the last biosolid application was between 7 and 9 months. In addition, the application rate of biosolids strongly influenced topsoil concentrations of triclocarban. For example, in 2005 the average triclocarban concentration in fields with multiple biosolid applications was 131.9 ± 76.1 ng g^{-1} dry weight whereas for fields in receipt of a single biosolid application, the average triclocarban concentration was 107.1 ± 43.7 ng g^{-1} dry weight. Nevertheless, triclocarban remained present in agricultural soils years after the last biosolids application, with concentrations reaching a

maximum of 45.8 ± 6.1 and 72.4 ± 15.3 ng g^{-1} dry weight, 7 and 8 years respectively after the last biosolids application.

The persistence of triclocarban in biosolid-amended soils was also reported by Thelusmond et al. (2018) with dissipation half-lives reported to range between 165 – 190 days under aerobic conditions. Whilst the reported half-lives for carbamazepine in four agricultural soils were similar and ranged between 128 – 241 days, diclofenac was rapidly removed within one week. Results suggested that soil organic carbon content has an important role in the fate of pharmaceuticals in soils. Soils with higher organic carbon matter appeared to sorb the chemical and thereby reducing the potential for biodegradation as diclofenac was depleted in the soils with lower organic matter (1.4, 1.9%), but was still present in soils with higher organic carbon matter (2.8, 2.4%) at day 3. Comparatively soils with a lower organic matter content exhibited lower carbamazepine concentrations at days 21 and 40.

Shao et al. (2018) observed much longer half-lives for carbamazepine in composted sewage sludge amended agricultural soils than reported by Thelusmond et al. (2018). Depending on the rate of sludge application, carbamazepine half-lives ranged from 533.19 – 1386.29 days and increased with increasing sludge addition rate. While the half-lives of triclosan were much smaller than for carbamazepine, similar results were observed in terms of increasing percent addition of composted sewage sludge resulted in increased the half-lives of triclosan in the agricultural soil mesocosms (4.85 - 55.45 days). In a similar conclusion to Thelusmond et al. (2018), Shao et al. (2018) highlighted the role of total organic carbon on contaminant dissipation by observing a significant positive correlation between organic content and half-lives of triclosan and carbamazepine. Increasing the percent of composted sewage sludge in the soil, increases the total organic carbon content of the soil which would promote sorption thereby decreasing the bioavailability of the chemicals and inhibiting the potential for degradation.

Seasonality was found to strongly influence the fate of pharmaceuticals in soils receiving treated wastewater irrigation Biel-Maeso et al. (2018b). Even though less irrigation occurred in winter months, higher concentrations of pharmaceuticals were detected in soils during this season between 10 and 14 ng/g in comparison to samples collected in the summer. It was expected that lower temperatures minimized biodegradation processes in soils in combination with lower irradiation levels in the winter months which decreased the potential for photodegradation. The vertical distribution of pharmaceuticals in soil columns was also influenced by the season as well as chemical properties of the pharmaceutical with more hydrophobic pharmaceuticals (> 4.5 log K_{ow}) observed at the highest concentrations in the soil columns (e.g. diclofenac and mefenamic acid) in the winter months. Nevertheless, periods of intense heavy rain strongly affected the vertical distribution of the chemical as pharmaceuticals were rapidly leached in the vadose zone demonstrating the significant role seasonal weather has on the fate of contaminants in soils.

In addition to these natural attenuation processes, research into remediation alternatives to reduce pharmaceuticals in soil environments has also emerged. One study exploring the removal of antibiotics from the environment was conducted by Wang et al. (2018a). They

explored the possibility of using various Fe-based metal-organic frameworks (MOFs) to help degrade tetracycline (of varying quantities) in the environment. They found that the main active species of the photocatalytic degradation process were ·O2-, ·OH and H+, which are used to assist in the breakdown of tetracycline to intermediate or end products. This process was found to be able to remove up to 96% of tetracycline present, with this percentage decreasing as the initial concentration increased.

The uptake of CECs into plants remains an area of active research especially with regards to antibiotic residues in soils. The presence of tetracyclines and sulphonamides in manure, soil and food crops were studied by Conde-Cid et al. (2018) with samples taken from two locations in Galicia, Spain. Antibiotics were found in each of these three matrices, in varying quantities and with a varying number of antibiotics in each. Overall, it could not be concluded that a high risk of soil contamination exists in the locations studied, although the concentrations of the antibiotics in crops were relatively high and suggests accumulation of these antibiotics in the plants.

The concentrations of eight tetracyclines taken up into grains (wheat and rye) were studied by Schwake-Anduschus and Langenkamper (2018) via hydroponic experiments and agricultural practice. It was found that the overall detection frequency for tetracyclines was 21%, along with conversion and degradation products also being detected. The levels of tetracyclines detected in the grains were high enough to select for antibiotic resistance in bacteria, although not high enough to be considered toxic to humans. Similarly, Sallach et al. (2018) investigated the uptake of three antibiotics (sulfamethoxazole, lincomycin and oxytetracycline) in lettuce grown in soils with increasing sand content and decreasing clay content. Differences in sorption behavior did not completely explain plant uptake as plant health and growth rates must also be accounted for. To this end, increased sand content lead to a higher toxic response, impacting the accumulation of the compounds in the tissues.

Reviews of antibiotics in natural environments and uptake into plants have been conducted by Grenni et al. (2018) and Madikizela et al. (2018), respectively. Grenni et al. (2018) review focuses on how the antibiotics in the environment can affect, either directly or indirectly, the microbial community by inhibiting some microbial groups involved in key ecosystem functions. They conclude by suggesting that in addition to controlling the use of antibiotics, studies to improve their degradation in natural environments are needed. This emphasizes the need to remove antibiotics from the environment. Madikizela et al. (2018) reviewed work conducted on the uptake of antibiotics, including the challenges in the detection and uptake of the antibiotics as well as the physicochemical properties influencing plant uptake. The review identified a lack of real-life situations studied, such as the occurrence of pharmaceuticals in vegetables bought in supermarkets. The authors suggest that investigations into the effects of extended daily consumption of pharmaceuticals through contaminated food and water should be conducted.

ANTHELMINTICS

New studies have investigated the occurrence of anthelmintic compounds in wastewater treatment plants (WWTPs), including D'Alessio et al. (2018, Wang et al. (2018b) as well as in surface and groundwater Sodré et al. (2018, Yang et al. (2018c). For example, D'Alessio D'Alessio et al. (2018) investigating the occurrence and removal of 43 pharmaceuticals and steroid compounds at four different WWTPs in Hawai'i as well as their environmental fate, observed thiabendazole, TBZ, in the raw as well as in the treated wastewater samples at concentrations below the analytical detection limit D'Alessio et al. (2018). Wang et al. (2018b) investigated the occurrence of TBZ in seven WWTPs in Xiamen, southeast China. Average mass load of TBZ in the influent and effluent samples was 0.210 and 0.280 g/d, respectively, with an average removal efficiency of -23% and a mass loss proportion of -49 % Wang et al. (2018b). Negative values may be due to measurement uncertainty, concentration fluctuations caused by the sample collection and hydraulic retention time, compounds released from the adsorbed phase, lack of the degrading bacteria in the activated sludge, and analytes transformed from the conjugated forms Wang et al. (2018b).

Sodré et al. (2018) investigated the occurrence of different classes of emerging micropollutants, in Brazilian surface waters from 1999 to 2018. Mebendazole was the only anthelimintic compound investigated, and ten samples, were collected. Only one sample showed a concentration of mebendazole (14 ng/L) higher than the analytical detection limit.

Yang et al. (2018c) investigated the occurrence and distribution of 93 pharmaceuticals and personal care products, including TBZ, in surface water and groundwater of Dongjiang River basin in south China. The concentration levels of TBZ was relatively low (median concentration < 1 ng/L).

While the occurrence of anthelmintic compounds in the environment has been investigated during the past few years Kim et al. (2017, Kumirska et al. (2016, Yang et al. (2018c), limited to no information are available on their environmental fate. In an attempt to better understand and predict the environmental fate of these compounds, batch experiments have been conducted during the past year Babic et al. (2018, Mutavdzic Pavlovic et al. (2018, Tran et al. (2018).

For example, Babic et al. (2018) investigated for the first time abiotic elimination processes (hydrolysis, photolysis, and sorption) of anthelminthic drug febantel, the occurrence of its metabolic products, and its environmental fate. Febantel was persistent against solar radiation. Kinetics of hydrolytic elimination was highly impacted by pH, ionic strength, and temperature with half-lives ranging between 210 min and 99 days. Fenbendazole (FBZ) and fenbendazole sulfone represented the major degradation products. Catalyzed hydrolysis followed by consecutive oxidative cyclization to the five-membered ring of the benzo-imidazole derivative represented the hydrolytic degradation pathway. Sorption capacity of febantel increased with decreasing pH value and ionic strength of febantel solution. The organic carbon-normalized coefficient, K_{OC}, ranged between 1490 and 3894 mg/L for the sediment samples. A

pseudo-second-order kinetic model best described the rate of febantel sorption Babic et al. (2018).

Mutavdzic Pavlovic et al. (2018) investigated the sorption process of albendazole (ALB) on five sediment samples and five soil samples from Croatia's region with different physio-chemical properties as well as the impact of contact time, initial concentration, ionic strength and pH. The estimated sorption coefficient, K_d, ranged between 29 and 104 mg/L depending on the sediment or soil characteristics. Sorption capacity of ALB decreased with an increase in initial concentration and contact time. K_d values were the highest at the lowest pH tested (pH 5.0), and decreased as pH increased (up to pH 8.0) due to the ionization states of ALB. In contrast with pH, sorption of ALB decreased with an increase in ionic strength. Organic matter, content of copper and zinc, and pH were the major factors influencing sorption of ALB. A pseudo-second-order kinetic model best described the rate of ALB sorption.

Ljubas et al. (2018) investigated the degradation of ALB in the presence of four UV-based advanced oxidation processes such as UV photolysis, UV photocatalysis (over TiO_2 film), UV and ozone, and UV and H_2O_2. The fastest and slowest degradation of ALB and its degradation products were obtained in the presence of UV and H_2O_2, and UV photolysis, respectively.

Liou and Chen (2018) identified ABZ transformation products and elucidated the pathways and mechanisms that result from its reaction with MnO_2 in the presence of different levels of pH, metal cations (e.g., Ca^{2+} and Mg^{2+}) and organic matter. Minor effects from ionic strength and metal cations on ABZ degradation were observed. By contrast, decrease of pH greatly enhanced the reaction rate. Surface complexation between ABZ and MnO_2 was indicated to be the dominant control in the reaction kinetics. Humic acid was found to cause significant inhibition due to the reductive dissolution of MnO2.

Tran et al. (2018) investigated the adsorption and transformation of the anthelmintic drug niclosamide, NIS, by a synthetic suspension of birnessite that is similar to natural MnO_2. The impact of different levels of birnessite, pH and organic matter was also investigated. The manganese oxide birnessite adsorbed and catalyzed the transformation of NIS into 2-chloro-4-nitroaniline and 5-chlorosalicylic acid at acidic pH. Adsorption and rate of transformation decreased when pH increased from 4.0 to 5.5. In the presence of natural organic matter, the transformation rates of NIS decreased because of competition for reactive sites and interaction between NIS and organic matter in solution and/or at the surface of MnO_2. Biodegradation and adsorption represented the predominant removal mechanisms in the presence of natural occurring environmental conditions.

Stuchlikova et al. (2018) investigated the metabolic pathways of two anthelmintic compounds, FBZ and flubendazole (FLU) in the presence one of the most commonly occurring plants in pastures, ribwort plantain (*Plantago lanceolate L.*). The enzymatic system of the ribwort plantain was able to uptake FLU and FBZ, translocate them in leaves and transform them into several metabolites, particularly glycosides. In addition, 12 FLU and 22 FBZ metabolites were identified in the root, leaf base and leaf top of the plant. However, most of FLU and FBZ

metabolites forms in the ribwort was decomposed back to the biologically active parent drug. Both anthelmintic compounds in the ribwort plantain cells caused significant increase of proline concentration (up to twice), a well-known stress marker, and significant decrease of superoxide dismutase activity (by 50%).

Papadopoulou et al. (2018) addressed the bioaugmentation potential of a natural isolated TBZ-degrading bacterial consortium in a natural contaminated soil having a natural gradient of TBZ levels ranging between 12 and 12000 mg/kg, as well as in a soil with similar physicochemical properties and soil microbiota, which was artificially, contaminated with the same TBZ levels. Removal of TBZ in the bioaugmented soils was concentration dependent. In fact, bioaugmentation effectively removed TBZ from both soils at levels up to 400 mg/kg but failed at the highest contamination level (12000 mg/kg). Bioaugmentation had no effect on the soil bacterial diversity, while long-term contamination with extremely high levels of TBZ drastically reduced bacterial diversity, and dominance of γ- and β-proteobacteria.

MICROPLASTICS

Few topics have received more attention in recent years than that of microplastics (MP). While the majority of MP research has focused on marine systems, measuring the occurrence and effects of MP in the terrestrial environment began to emerge in 2018. Three reviews (Chae and An (2018, He et al. (2018, Ng et al. (2018)) frame the current state of knowledge. Common trends emerge from these reviews including the need for standardization of methods for the extraction, quantification, and identification of MP materials. Focus is needed on a wide range of soil organisms, as earthworms are the most common model organisms for toxicity studies. As 'microplastics' represent a wide range of materials, characterization and toxicity testing representing this range of material is necessary. Ng et al. highlight the need for studies on MP bioavailability and He et al. identify the need for information related to source and fate of MP in terrestrial ecosystems. Chae and An recommend consideration on the chemical additives that are associated with plastic products including plasticizers, retardant, antioxidants, and photo-stabilizers.

Wang et al. (2018c) demonstrated the need for improved methods for the extraction of MPs from complex terrestrial matrices. Using flotation and H2O2 digestion, Wang and colleagues showed that 100 μm MP beads could be completely recovered from soil and biosolids, while smaller (0.05-4.8 μm) MP bead recoveries ranged from 5-80 %, with an average of 20 %. A multi-step extraction with H20 and ZnCl2 improved recovery in MP beads > 1μm, but recovery of the smallest MP beds remained low. The authors acknowledge that separation from organic matter is necessary for MP bead extraction, but the common H202 method results in poor extraction efficiencies.

Piehl et al. (2018) employed FTIR to quantify macro- and MP contamination in agricultural farmland in Germany. Polyethylene MPs was most commonly detected followed by polystyrene and polypropylene. Concentrations of 0.34 particles per kg dry weight of soil were measured at this site which had never received microplastic-containing fertilizer or used agricultural plastic applications. Contamination of other soils receiving mulch, silage films,

or plastic containing fertilizers like sewage sludge or biowaste components were theorized to be much higher.

Similarly, Liu et al. (2018) measured MP contamination in a Chinese farmland. Concentrations of 78 particles per kg were measured in top soil with polypropylene (50.51 %) and polyethylene (43.43 %) dominating.

Four common MP types including polyacrylic fibers, polyamide beads, polyester fibers, and polyethylene fragments were introduced to soils in a study by de Souza Machado et al. (2018). They showed that this exposure affected soil bulk density, water holding capacity, and the functional relationship between microbial activity and water stable aggregation in a concentration dependent manner. This result highlights both the need for quantification rather than purely qualitative MP data and the fact that MP contamination represents a relevant long-term anthropogenic stressor in terrestrial ecoystems.

Exposure of 250-1000 μm polyethylene MPs at concentrations ranging from 62-1000 mg/kg soil was investigated by Rodriguez-Seijo et al. (2018). Using a combination of Fourier transform infrared spectrometry (FTIR) and nuclear magnetic resonance (NMR), molecular biomarkers for oxidative stress and energy metabolism were evaluated in earthworms. Results showed that a complex toxic response was observed at concentrations exceeding 250 mg/kg soil.

The impact of earthworms on the effects of MPs emanating from plastic mulch soil additives on the soil-plant system was studied by Qi et al. (2018). Qi and colleagues evaluated the impacts of 1 % concentrations of MP residues from either a polyethylene or a starch based biodegradable plastic mulch on the growth of wheat. Results showed that the biodegradable starch-based residue had a significantly greater negative impact on wheat growth. However, the addition of earthworms into the growth systems alleviated these impairments.

In agricultural systems, contamination occurs in mixtures of contaminants of different types. While this dynamic makes toxicity studies challenging, their importance was exemplified in the work of Yang et al. (2018b). Yang and co-authors examined the impact of micrometer MP contamination on the decay of the herbicide glyphosate. While MPs had little effect on glyphosate decay, they did impact soil microbial respiration and the dynamics of soil β-glucosidase, urease and phosphatase indicating their potential to threaten soil quality.

ENGINEERED NANOMATERIALS

Engineered nanomaterials (ENMs) have expanded its horizon together with technological advancements. This gain in prominence of ENMs is due to the enhancement of performance in physical, chemical and biological properties compared to their bulk counterparts Jeevanandam et al. (2018). This heavy usage of ENMs has implications in the environment. The latest threat is that of nanoplastics, which has been critically reviewed and its global presence has been realized Alimi et al. (2018).

Bundschuh et al. (2018) reviewed the recent advances in identifying the sources and fate of nanoparticles (NPs), summarized the effects of NPs in simplified studies, and presented how NPs interact with biota in a more

complex environment. However, the current studies acknowledge the presence of a wide gap between the increased classes of ENMs and its application versus their toxicity assessment Matteis and Rinaldi (2018). In the recent times, considerable efforts has been put to understand life cycle of ENMs and studies have recognized ENMs fate and transformation pathway in the environment Pourzahedi et al. (2018).

In the context of agricultural ecosystems publications centering ENMs has seen an increase in 2018, which is understandable as nanotechnology usage in enhancing agriculture productivity is on rise Kah et al. (2018, Moore and Gardea-Torresdey (2018). The present focus is on understanding the impact of ENMs exposure on plant and soil, which will be carried out by synthesizing recent research efforts.

Nanotechnology focus in agroecosystem is mainly in development of more efficient nano-based pesticides and fertilizers. Kah et al. analyzed 78 published work and determined that benefits from the usage of ENMs in agro-ecosystems is not tremendously high, with a median efficacy of 20-30% relative to conventional products Kah et al. (2018). Moreover, ENMs from other sources, which make their way to agricultural system still pose threat to crop quality and can impact soil health Falinski et al. (2018).

The recent identification of the fate, transformations and interactions of different ENMs in the agricultural soil environment has prioritized the studies toward the effects of their direct toxicity to plants. Inspecting the effect of nano-silicon (SiNP), a nanofertilizer, Asgari et al. (2018) found that SiNPs didn't have a toxic effect on the oat (*Avena sativa L.*) plants, but silicon transporter (*Lsi1*) gene was expressed in plants exposed to SiNPs, and there was also increase in phenylalanine ammonia lyase expression. Lignification in leaves and roots, were observed and SiNPs aggregated in the roots and deposited in the leaves.

Ogunkunle and co-authors examined the uptake and consequent toxicity of manufactured nano-copper in cowpea (*Vigna unguiculata*) grown on soil substrate. Their study indicated that copper NPs bioaccumulated in the leaf and cowpea seeds. Seeds accumulated around 500 mg/kg for <25 nm sized particle and around 1000 mg/kg for 60-80 nm particles. The induced nanotoxicity enhanced activity of ascorbate peroxidase, catalase and glutathione reductase Ogunkunle et al. (2018).

Simonin et al. found that copper oxide (CuO) NP have detrimental effect on microbial activity in soil, with contrasting physicochemical properties, impacting carbon and nitrogen cycle. In a 90 days experiment comprising of five different agricultural soil types, with varied concentration of CuO NPs, it was observed that response to denitrification was most sensitive. Heterotrophic microbial activities decreased substantially in soils planted with wheat, even at low concentrations of CuO NPs. Also, the negative effects of CuO NPs increased over time Simonin et al. (2018).

Zinc oxide (ZnO) NPs are also known to be harmful to soil microbial community and it can adversely affect microbial enzymatic activities Garcia-Gomez et al. (2018a). ZnO NPs phytotoxicity was also evaluated in nine different crops *viz,* wheat, maize, radish, bean, lettuce,

tomato, pea, cucumber, and beet in pot experiment Garcia-Gomez et al. (2018b). The study concluded that Zn phytotoxicity was dependent of the soil pH; cucumber and beet were the most sensitive species to ZnO NP in the calcareous soil Garcia-Gomez et al. (2018b).

Another oxide ENM, cerium oxide (CeO_2), which is among the top 10 NPs produced worldwide Ma et al. (2018b), generally remains strongly absorbed in agricultural soil, but may undergo reductive dissolution releasing Ce(III) Arai and Dahle (2018). This strong adsorption of CeO_2 NPs restricts its uptake and is generally limited to roots, but reduction to Ce(III) may enhance the uptake of ceria in barleyRico et al. (2018). In another study by Ma et al. on CeO_2 NPs uptake by lettuce (*L. sativa*) in pot experiment, it was found that under root exposure of NPs, concentrations of Ce for soil amended with 500 and 1000 mg of CeO_2 NPs/kg were, roots: 44.5 ± 20.7; leaves: 0.09 ± 0.04 and roots: 79.5±62.0; leaves: 0.3±0.17 μg/g, respectively. Majority of CeO_2 NPs was accumulated in the roots, and the Ce contents of the shoots were at least 2 orders of magnitude lower than that in the roots. In the same study of Ma et al., under foliar exposure of CeO_2, Ce concentrations in the treated leaves (exposed to CeO_2), untreated leaves (without CeO2), and roots of lettuce were 982.4 ± 341.7, 8.1 ± 3.4, and 2.7 ± 0.4 μg/g, most of the CeO_2 was confined in the leaves, where the exposure was maximum {Ma, 2018 #7605}.

One of the main hurdles of studying ENMs and nanotoxicity is its easy detection in different environmental systems. Keller and co-authors developed methods for detection of nano-copper using single-particle ICP-MS in edible tissues Keller et al. (2018), these kind of analytical studies for detecting NPs in food crops is very much needed.

Ma et al., reviewed the uptake of ENMs by food crops and proposed that to optimize the safe and sustainable application of nanotechnology in agroecosytem both advantages and disadvantages of ENMs needs to be understood Ma et al. (2018a). Achari and Kowshik also resonated similar propositions as Ma et al., on the recent developments of nanotechnology usage in agriculture Achari and Kowshik (2018), they further stressed that more field study is needed for through risk assessment of ENMs and to understand the toxicological impact of ENMs in the true environmental setup.

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