

VIRGINIA WATER RESOURCES RESEARCH CENTER

Emerging Contaminants in the Waters of Virginia

**2019 Report of the Academic Advisory Committee for Virginia Department of
Environmental Quality**



SPECIAL REPORT



**VIRGINIA POLYTECHNIC INSTITUTE AND STATE UNIVERSITY
BLACKSBURG, VIRGINIA**

**SR63-2019
October 2019**

This special report is a publication of the Virginia Water Resources Research Center. The research was supported with funds provided by the Virginia Department of Environmental Quality. The views expressed are those of the individual authors and do not necessarily reflect the views or policies of the Virginia Department of Environmental Quality or the Virginia Water Resources Research Center. The mention of commercial products, trade names, or services does not constitute an endorsement or recommendation.

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EMERGING CONTAMINANTS IN THE WATERS OF VIRGINIA

**2019 Report of the Academic Advisory Committee for
Virginia Department of Environmental Quality**

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**Publication of the
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**SR63-2019
October 2019**

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Acronyms, Abbreviations, and Symbols

Throughout this report, chemical concentration units have been converted to parts per million (ppm), parts per billion (ppb), parts per trillion (ppt), or parts per quadrillion (ppq) in the interest of consistency and ease of reading.

µm – micrometer = one millionth of a meter = 1/1,000,000 meter

% – percent = 1/100

∑ – sum of

\$ – United States dollar

AAC – Academic Advisory Committee

AAPS – American Association of Pharmaceutical Scientists

AFFF – aqueous film-forming foams

ARE – antimicrobial-resistant elements

ARG – antimicrobial-resistant genes

ARM – antimicrobial-resistant microorganisms

ATSDR – Agency for Toxic Substances and Disease Registry

BAF – bioaccumulation factor = The net accumulation of a chemical by an aquatic organism as a result of uptake from all environmental sources (e.g., water, food, sediment, soil). It is the ratio of the concentration of a contaminant in an organism to its concentration in the surrounding environment (e.g., water, sediment, soil).

BCF – bioconcentration factor = The net accumulation of a chemical by an aquatic organism as a result of uptake directly from the ambient water, through gill membranes or other external body surfaces. It is the ratio of the concentration of a contaminant in an organism to its concentration in the surrounding water.

BTBPE – 1,2-bis(2,4,6-tribromophenoxy) ethane

CAFO – concentrated animal feeding operation

CBW – Chesapeake Bay watershed

CCL 4 – Contaminant Candidate List 4 = Currently not subject to any proposed or promulgated national primary drinking water regulations but are known or anticipated to occur in public water systems. EPA announced the Final CCL 4 on November 17, 2016.

CDC – United States Centers for Disease Control and Prevention

CEC – contaminant of emerging concern

cm – centimeter = 1/100 of a meter

CNT – carbon nanotube

DBDPE – decabromodiphenyl ethane

DEQ – Virginia Department of Environmental Quality

d.w. – dry weight

EDC – endocrine-disrupting chemical

EE2 – ethynylestradiol

e.g. – Latin *exempli gratia* = “for example”

ENM – engineered nanomaterial

EPA – United States Environmental Protection Agency

etc. – Latin *et cetera* = “and so forth”

FAA – United States Federal Aviation Administration

FDA – United States Food and Drug Administration

FY – fiscal year
 GenX chemicals – used as replacements for PFOA
 HBB – hexabromobenzene
 HBCD – hexabromocyclododecane
 HFPO – hexafluoropropylene oxide
 HPV – high production volume
 i.e. – Latin *id est* = “that is”
 kg – kilogram = 1,000 grams
 km – kilometer = 1,000 meters
 K_{oc} – soil organic carbon-water partition coefficient = The ratio of the concentration of a chemical adsorbed per unit mass of soil organic carbon to its concentration in water. It is usually expressed as log K_{oc}; $K_{oc} = (K_d * 100) / \text{organic carbon content in a soil (\% weight basis)}$, where the soil/water partition coefficient of a chemical (K_d) = concentration of a chemical in soil/concentration of a chemical in water.
 K_{ow} – octanol-water partition coefficient; K_{ow} = concentration in octanol/concentration in water. It indicates the tendency of an organic compound to adsorb to soil and living organisms. It is usually expressed as log K_{ow}; log K_{ow} tends to be inversely related to water solubility and directly proportional to the molecular weight of a substance.
 L – liter
 L/g – liter per gram
 L/kg – liter per kilogram
 LOQ – limit of quantification
 l.w. – lipid weight
 MCL – maximum contaminant level
 MeEE2 – mestranol
 mg – milligram = one thousandth of a gram = 1/1,000 gram
 mg/kg – milligram per kilogram = 1 part per million
 mg/L – milligram per liter = 1 part per million
 mm – millimeter = one thousandth of a meter = 1/1,000 meter
 MRL – minimum reporting level
 NCCOS – National Centers for Coastal Ocean Science
 ng/kg – nanogram per kilogram = 1 part per trillion
 ng/L – nanogram per liter = 1 part per trillion
 nm – nanometer = one billionth of a meter = 1/1,000,000,000 meter
 NOAA – National Oceanic and Atmospheric Administration
 NOS – National Ocean Service
 NQ – not quantifiable
 OTC – over the counter = Medicines that do not require a prescription.
 PBDE – polybrominated diphenyl ethers
 PBEB – pentabromoethylbenzene
 PFAS – per- and polyfluoroalkyl substances (C_nF_(2n+1)-R)
 PFBS – perfluorobutane sulfonic acid
 PFC – perfluorinated compounds
 PFCA – perfluoroalkyl carboxylic acids
 PFDA – perfluorodecanoic acid
 PFDS – perfluorodecane sulfonic acid

PFHpA – perfluoroheptanoic acid
 PFHxA – perfluorohexanoic acid
 PFHxS – perfluorohexane sulfonic acid
 PFNA – perfluorononanoic acid
 PFOA – perfluorooctanoic acid
 PFOS – perfluorooctanesulfonate
 PFSA – perfluoroalkane sulfonic acids
 PFTeDA – perfluorotetradecanoate
 pg – picogram = one trillionth of a gram = 1/1,000,000,000,000 gram
 pg/kg – picogram per kilogram = parts per quadrillion
 pg/L – picogram per liter = parts per quadrillion
 pH – Describes the acidity or alkalinity of a solution on a logarithmic scale on which 7 is neutral, lower values are more acidic, and higher values more alkaline.
 pK_a – acid dissociation constant = The negative log of the acid dissociation constant (K_a) value. The more positive the value of pK_a, the smaller the extent of dissociation at any given pH (Henderson-Hasselbalch equation) – that is, the weaker the acid. A weak acid has a pK_a value in the approximate range of -2 to 12 in water.
 POP – persistent organic pollutant
 ppb – parts per billion = 1/1,000,000,000 = μg/L or μg/kg
 PPCPs – pharmaceuticals and personal care products
 ppm – parts per million = 1/1,000,000 = mg/L or mg/kg
 ppq – parts per quadrillion = 1/1,000,000,000,000,000 = pg/L or pg/kg
 ppt – parts per trillion = 1/1,000,000,000,000 = ng/L or ng/kg
 t/ha – tons per hectare
 UCMR 3 – Third EPA Unregulated Contaminant Monitoring Rule
 U.S. – United States
 USAF – United States Air Force
 USDA – United States Department of Agriculture
 USGS – United States Geological Survey
 w.w. – wet weight
 WWTP – wastewater treatment plant

Introduction

This report details the activities and findings of the Academic Advisory Committee (ACC) to the Virginia Department of Environmental Quality (DEQ) for Section 3 of the fiscal year (FY) 2019 approved scope of work. The scope of work calls for “a literature review to inform a future monitoring strategy to assess emerging contaminants in Virginia waters.” The DEQ is considering the need to expand its monitoring strategy to evaluate the presence, magnitude, risks of, and potential abatement strategies for emerging contaminants in the waters of Virginia. To initiate work towards this endeavor, the AAC conducted a literature review to address the interest expressed by DEQ in emerging contaminants in the waters of Virginia.

This literature review builds upon the document *Toxic Contaminants in the Chesapeake Bay and its Watershed: Extent and Severity of Occurrence and Potential Biological Effects – 2012 Technical Report*.¹ All contaminant groups of emerging concern for the waters of Virginia (i.e., all for which up-to-date peer-reviewed journal articles or official government reports are found) are listed. This literature review covers a wide list of emerging contaminants in detail, including:

- Pharmaceuticals and personal care products (PPCPs)
- Flame retardants
- Hormones and endocrine disruptors
- Perfluorinated compounds
- Antibiotics, antibiotic-resistant microorganisms, and antibiotic-resistant genes
- Micro- and nano-plastics and microfibers
- Engineered nanomaterials

For each of the above listed categories, the major sources, occurrences, environmental and human health concerns, and environmental behaviors and pathways are reviewed.

1. Emerging Contaminants – An Overview

Emerging contaminants are defined by the United States Environmental Protection Agency (EPA) as “contaminants of emerging concern (CECs).”² The EPA definition of CECs further states that:

- They include chemicals and other substances that have no regulatory standard.
- They have been recently “discovered” in natural streams (often because of improved analytical chemistry detection levels) and may cause deleterious effects in aquatic life at environmentally relevant concentrations.
- They are pollutants not currently included in routine monitoring programs and may be candidates for future regulation depending on their (eco)toxicity, potential health effects, public perception, and frequency of occurrence in environmental media.
- They are not necessarily new chemicals.
- They include pollutants that have often been present in the environment, but whose presence and significance are only now being evaluated.

Since the 2002 landmark investigation by the United States Geological Survey (USGS) that reported widespread detection of CECs in the streams of the United States (U.S.),³ scientific reports related to CECs have increased exponentially, and public concerns on their potential human and environmental impacts are on the rise. The list of CECs has since expanded to include not only persistent organic pollutants (POPs), PPCPs, veterinary medicines, endocrine-disrupting chemicals (EDCs), and nanomaterials, but also microplastics and microfibers.⁴⁻⁶ As shown in Figure 1, anthropogenic activities are the source and cause of CECs in the environment. Contaminants of emerging concern enter aquatic systems through numerous pathways, of which wastewater treatment plants (WWTPs), concentrated animal feeding operations (CAFOs), and land application of animal manure and biosolids are considered major critical control points.⁷⁻⁹ Additionally, recent studies have suggested that aging sewer distribution systems as well as stormwater overflow and runoff could be important routes for CECs to enter the aquatic environment.^{10, 11} Furthermore, specific usage patterns of certain categories of CECs result in specific routes of entry to the environment.

A recent comprehensive literature review¹² of CECs reported widespread detection in a range of environmental matrices from 71 countries covering all continents. The review included 1,016 original publications and 150 review articles on CECs (including PPCPs, hormones, and antibiotics) and their transformation products (new chemical compounds that form from a parent compound during wastewater treatment; transformation products have different properties than the parent compound). In total, this review reported 631 compounds in different environmental matrices at concentrations ranging from parts per trillion (ppt) to parts per million (ppm).

The detection of flame retardants, many of which are endocrine disruptors, within various environmental matrices has appeared frequently in the scientific literature and reports since the 1980s. Their concentrations are reported in ppt in water and in parts per billion (ppb) in sediment and in aquatic organisms (concentrations of flame retardants are 1,000 times greater in sediments and aquatic organisms than in water).^{13, 14} Perfluorinated compounds, another class of CECs, have increasingly gained attention because of their occurrence in natural waters; and although reported at low ppt levels,¹⁵⁻¹⁷ new evidence suggests potential negative impacts on human health

even at these low levels.¹⁸ Worldwide attention on microplastics, microfibers, and nanomaterials initially focused on their occurrence in marine environments but has expanded recently to freshwater, terrestrial, and atmospheric systems.^{19, 20} Their environmental occurrence has been reported in freshwater and marine environments globally.²¹⁻²³

Currently, there are no existing benchmarks or water-quality standards for CECs. In 2008, EPA developed a white paper to address the challenges and recommendations for assessing aquatic life criteria for CECs.² In this EPA document, endocrine disruptors received the most attention because of their significant adverse impact on natural populations of aquatic vertebrates at levels as low as ppt.²⁴⁻²⁶ Among the 97 chemicals listed on the EPA Contaminant Candidate List (CCL 4) for public drinking water systems, there are nine endocrine disruptors (three natural hormones, four synthetic hormones, and two other endocrine disruptor chemicals), one antibiotic compound, and two perfluorinated compounds.²⁷

In summary, CECs are frequently reported to occur in different environmental matrices in the U.S. and worldwide. For many of the CECs, the primary sources have been identified. However, there is a lack of systematic information on their sources and occurrences in the waters of Virginia. This information is essential for informing future needs and strategies of evaluating the presence, magnitude, and risks of CECs in Virginia. Hence, major sources, occurrences, environmental behaviors, and pathways of seven CEC categories are presented in the subsequent sections of this report with the goal of providing comprehensive and current information on CECs in the waters of Virginia.

The document *Toxic Contaminants in the Chesapeake Bay and its Watershed: Extent and Severity of Occurrence and Potential Biological Effects – 2012 Technical Report*¹ concludes that owing to the limitations of the data, there is uncertainty on the extent of occurrence and severity of PPCPs, flame retardants, and biogenic hormones in the Chesapeake Bay watershed (CBW). The major research gaps identified in the report are the limited understanding of the relationships among the sources of these contaminants, their pathways into the environment, and exposures to organisms. Literature published since 2012, additional information on Virginia waterbodies that are not in the CBW, and CECs that were not listed in the 2012 Technical Report are described in this review.

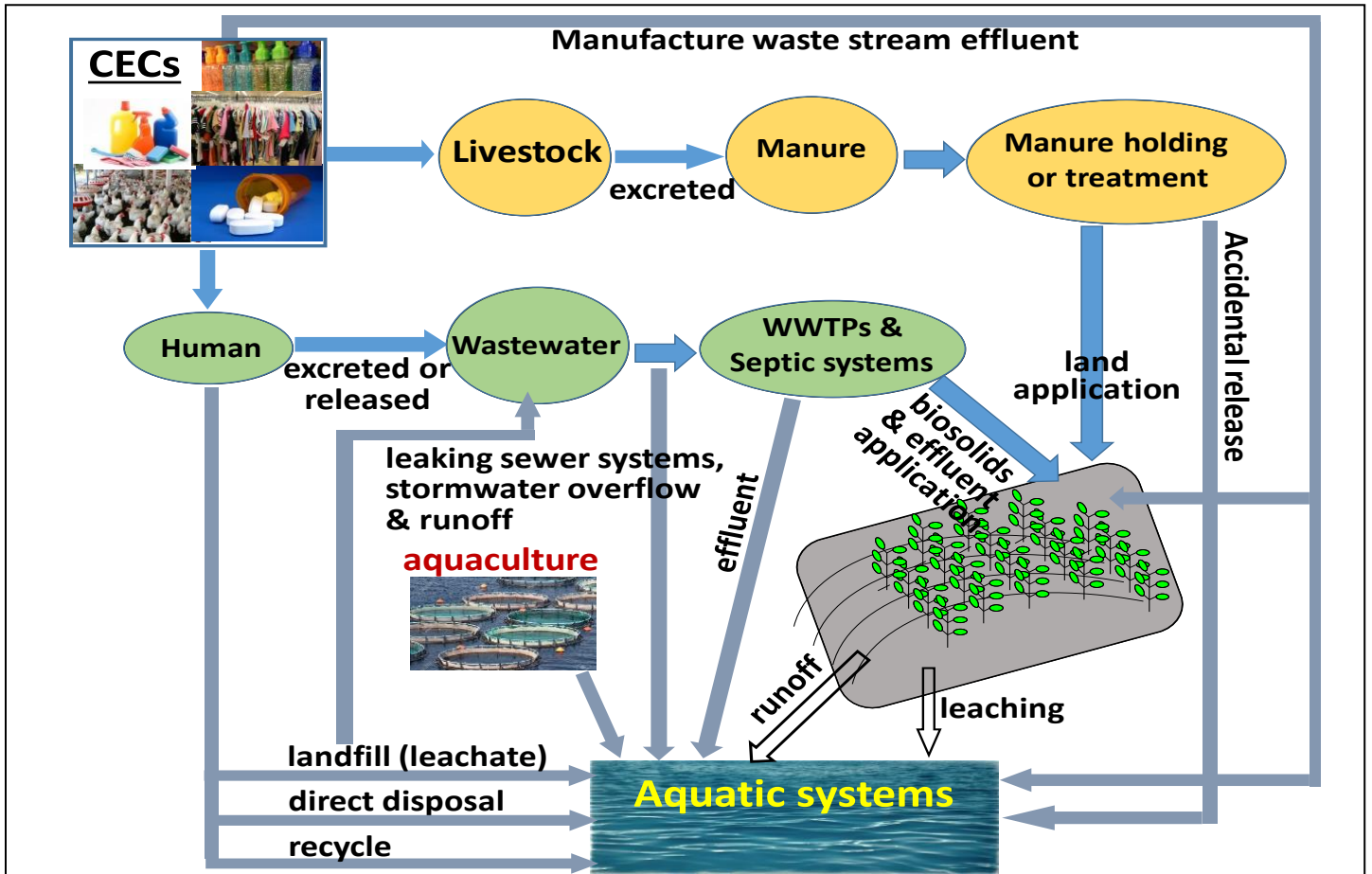


Figure 1. Major sources and pathways of contaminants of emerging concern (CECs) in the environment. Some CECs (e.g., plastic microfibers) can also disperse through the atmosphere.

2. Pharmaceuticals and Personal Care Products (PPCPs)

2.1. Major Sources

Each year, large quantities of pharmaceuticals are sold and consumed in the U.S. and worldwide for the diagnosis, treatment, or prevention of human illness and diseases. Worldwide, total prescription drug sales were \$811 billion in 2018. They are predicted to increase 6.5% annually and reach \$1.2 trillion in 2024, of which 41% of sales are expected to be in the U.S.²⁸ In 2017, 3.7 billion prescriptions were ordered or provided in the U.S.; the most frequently prescribed therapeutic classes are analgesics (painkillers), antihyperlipidemic agents (cholesterol-lowering drugs), and antidepressants.²⁹ Although over-the-counter (OTC) nonprescription drug use is more difficult to track, about 60% of pharmaceuticals used in the U.S. are OTC drugs²⁹ for pain relief, colds, flu, allergies, etc. In Virginia, about 93.3 million drug prescriptions were filled in 2018.³⁰ Based on the national average, it is reasonable to estimate that about 140 million nonprescription drug purchases are occurring yearly in Virginia. In addition to human usage, in 2018 about \$10.2 billion were spent on prescription and OTC pet medications for the care of companion animals, which are owned by about 68% of U.S. households.^{31, 32} Furthermore, large quantities (included in the EPA list of high-production-volume, HPV, chemicals) of personal care products, such as food supplements, fragrances, skin care and hair care products, insect repellents, and cleaning products are produced and sold in the U.S. each year.³³ Because antibiotics are used extensively in agricultural animal productions, this topic is discussed separately in Section 6 of this document.

Most pharmaceutical compounds are designed to be non-bioaccumulative (they do not become concentrated inside the body), with 50-95% typically eliminated as urine from the human body shortly after administration.³⁴ Ingredients in many household personal care products are released immediately after use. Collective and frequent use of PPCPs results in their continuous input to sewer systems leading to WWTPs or to onsite sewage disposal such as septic tanks. Research has shown that depending on the chemical characteristics of PPCPs, removal efficiencies from water at WWTPs varies from non-removal to 100% removal of the original compound.³⁵ However, 100% removal of the original compound from water does not indicate complete absence of its components. The removal of a compound from wastewater is often merely a transfer process during which a compound partitions from the aqueous phase into the solid phase during the wastewater treatment processes or transforms into intermediate products that may be more biologically active than the parent compound.³⁶ Therefore, WWTP effluent and biosolids can be significant sources of PPCPs. Similarly, septic system discharges have been identified as another important source of PPCPs.³⁷ A 2012 survey estimated that municipal WWTPs throughout Virginia treated approximately 671 million gallons of wastewater per day and served more than 5.85 million Virginians.³⁸ The remaining 30% of the population in Virginia uses septic systems. In 2016, a total of 64,508 dry tons of biosolids were spread on approximately 43,000 acres of farmland and forestland in Virginia.³⁹ With increasing economic development and urban population growth in Virginia, the volume of WWTP effluent and the mass of biosolids generated is expected to increase, resulting in increased potential input of PPCPs to the waters of Virginia.

Effluents and biosolids discharged from WWTPs or septic systems can result in differing inputs of PPCPs to the environment based on the sources discharging into the treatment systems.

For example, owing to the intensive use of cleaning products and pharmaceuticals in commercial health care and hospital settings, concentrations of PPCPs associated with detergents and medical treatments are higher in the discharges of wastewater treatment systems receiving commercial and hospital wastewater than in domestic wastewater.⁴⁰⁻⁴² Currently, there are 103 inpatient and outpatient hospitals; 279 nursing facilities; 5,300 clinical laboratories; and 64 biotech, pharmaceutical and life sciences companies located throughout Virginia.⁴³ It is unclear how the discharges from those facilities affect the receiving WWTPs. Furthermore, aging urban sewer distribution systems, as well as stormwater overflow and runoff could be important routes for PPCPs entering the aquatic environment,¹⁰ especially for urban waters.⁴⁴

2.2. Occurrence

2.2.1. Aquatic Environment (Water, Sediment, and Organisms)

Thousands of publications have reported worldwide detection of hundreds of PPCPs in aquatic environments.¹² There have been limited investigations of PPCPs in the waters of Virginia, especially for the waters outside of the CBW (Figure 2). The numbers of detectable PPCPs and their concentration ranges in the waters of Virginia investigated since 2007 are comparable with what has been reported in areas outside Virginia.¹² A recent investigation in Virginia looked for 142 PPCPs in 57 private wells from two counties and six tap water samples from three counties outside the CBW; numerous PPCPs were detected, ranging from 2–20 compounds/well and 4–10 compounds/tap water sample. In contrast, a recent 2019 USGS survey of 103 PPCPs and 21 hormones in the groundwater of 60 sites collected from Virginia reported detection of only five PPCPs: methotrexate (~37 ppt), carbamazepine (124 ppt), lidocaine (40 ppt), and pseudoephedrine + ephedrine (15 ppt), each at one site, and meprobamate (~23 ppt) at two sites.⁴⁵

A recent investigation of water, sediment, oysters, and mussels from 14 sites at Chester River, Manokin River, Holland Straits, and Kitts Creek along the Lower Eastern Shore of the Chesapeake Bay in Maryland detected, for the first time, four UV-filter compounds. These compounds included benzophenone-3, 2-ethylhexyl-4-methoxycinnamate, homosalate, and octocrylene in all but one water sample at 7.6–188 ppt and in twelve sediment samples at 200 ppt to 74 ppb (dry weight, d.w.).⁴⁶ For the mussel and oyster samples tested in this study, benzophenone-3 was at 17–118 ppb (d.w.) for all sites; homosalate was at 6.7–158 ppb (d.w.); and octocrylene was at 3.3–20 ppb (d.w.) for thirteen sites. There was no detection of benzophenone-3 in the mussel and oyster samples.

Compared to information on water, information on sediment and aquatic organisms in the waters of Virginia is even more limited. An extensive review of the literature found reports of detectable PPCPs in sediment samples from only two Virginia waterbodies (north and south forks of the Shenandoah River) and in organisms from only one Virginia waterbody (James River; Figure 2). Again, the limited literature information found so far is comparable with what has been reported for locations outside of Virginia.^{12, 47}

2.2.2. Soil

Previous investigations have detected various PPCPs in biosolids at concentrations from ppb to ppm and in WWTP effluents from ppt to ppb.^{48, 49} Application of biosolids and WWTP

effluents to farmland introduces PPCPs to soil, resulting in their potential input to adjacent aquatic environments through surface runoff and leaching.⁵⁰⁻⁵² To date, it is unknown to what extent WWTP effluents are applied on land in Virginia. Considering about 46% of all Virginia biosolids are applied to approximately 43,000 acres of farmland and forestland in Virginia,³⁹ a substantial amount of PPCPs can potentially enter affected soil environments. However, information is lacking on the levels of PPCPs in biosolids applied to soils in Virginia. A study of a field in the Midwestern U.S. that extensively received biosolids showed accumulations of four PPCPs (triclosan, triclocarban, 4-nonylphenol, and polybrominated diphenyl ethers [PBDEs]) in soil up to 30 cm deep at concentrations ranging from low ppb to ppm (d.w.).⁵³ The level of one PPCP (triclosan) in the surface soil of farm fields in northern Virginia where biosolids were applied was reported to be 4.5–70 ppb (d.w.) depending on the application frequency and the time since application.⁵⁴ This level is about ten times less than the reported levels for this compound in biosolids-applied soils elsewhere.³⁴ The average level of triclosan in the biosolids used in the northern Virginia study was approximately 16 ppm (d.w.), which is well within the range of other reported levels of 90 ppb to 66 ppm (d.w.) for typical biosolids from conventional WWTPs in the U.S.⁴⁸

Although runoff and leaching of nutrients and microbial contaminants from fields in the mid-Atlantic region that received applications of biosolids have been well studied (and control practices have been recommended and implemented),⁵⁵⁻⁵⁸ there is a lack of assessment of runoff and leaching of PPCPs from fields in the region where biosolids and WWTP effluent have been applied. Research in Canada has detected a list of PPCPs in runoff at concentrations up to 1.3 ppb from fields receiving applications of biosolids; their concentrations ranged from 0.001% to 29% of the initial input to the soil from biosolids.^{50, 59} A later study in Colorado reported PPCPs in surface runoff (up to 10 ppb) from a field where biosolids had been applied at higher levels.⁶⁰ This study also reported that the combination of partitioning and degradation mechanisms of a PPCP compound affects its levels in the runoff.⁶⁰ A recent EPA report has indicated the need for more information in order to better assess the impact of PPCPs in the environment from lands receiving applications of biosolids.⁶¹

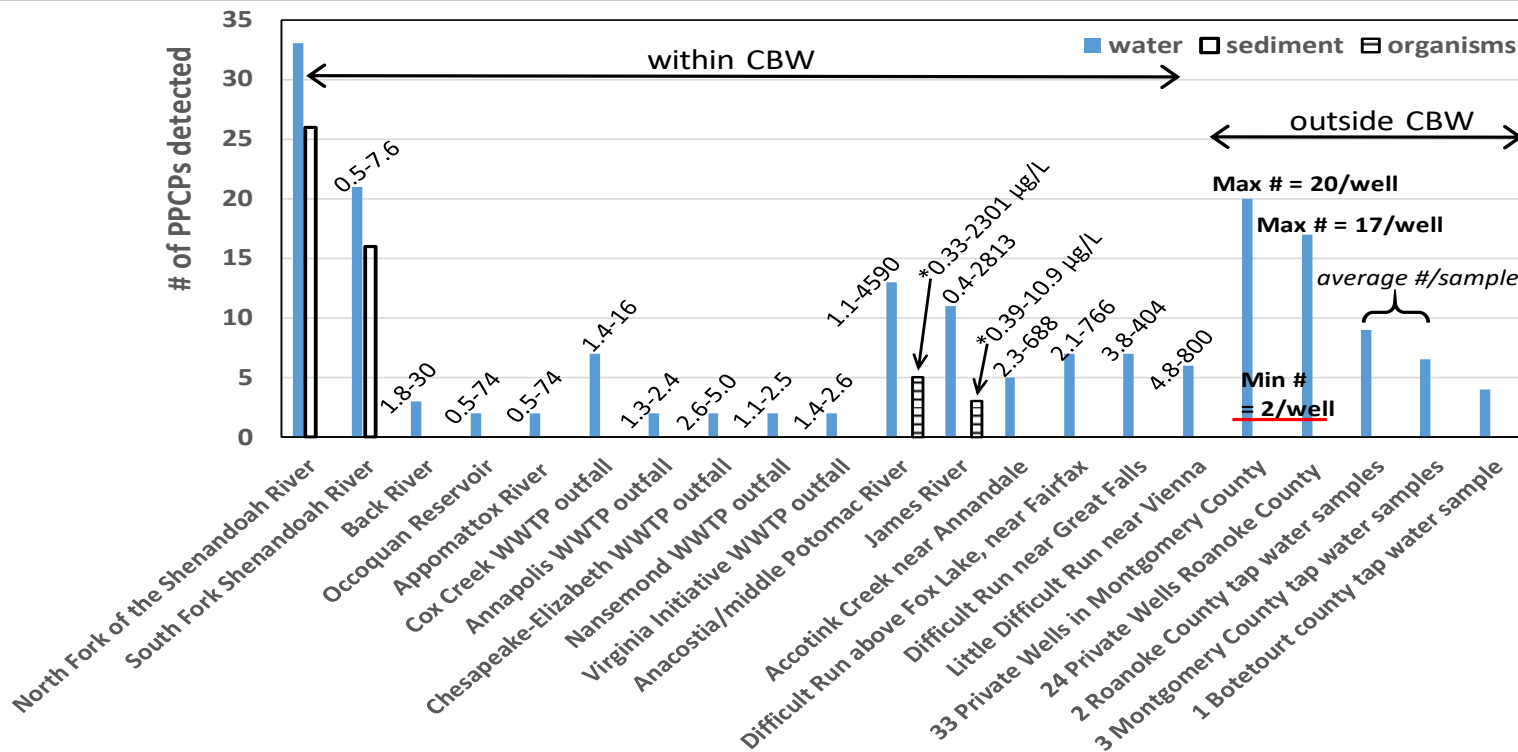


Figure 2. Number of PPCPs detected in Virginia, Maryland, and Washington, DC waters at selected locations. The numbers above a bar indicate the concentration range of detectable PPCPs at a sampling site of a waterbody. The concentration unit for the water samples is ppt (the numbers without the unit label). Asterisk mark indicates concentration range (ppb) of PPCPs in plasma of aquatic organisms tested (catfish, gizzard, shad, and carp). The minimum and maximum numbers of detectable PPCPs for the well waters in two Virginia counties are shown in bold font. The average number of detectable PPCPs per tap water from three Virginia counties is indicated in italic font. Table A1 of the Appendix includes detailed information pertaining to this figure. CBW = Chesapeake Bay watershed; µg/L = parts per billion (ppb)

3. Flame Retardants

3.1. Major Sources

Since the 1970s, flame retardants have been used in various products such as household furniture, textiles, plastics, and electronic equipment in order to reduce the risk of fire and meet fire safety regulations.⁶² Many consumer products contain flame retardants at 0.05–30% by weight.⁶³⁻⁶⁵

Among all flame retardants,⁶⁶ polybrominated diphenyl ethers (PBDEs), which include more than 200 potential congeners (variants or configurations of the chemical structure), have drawn extensive attention since the 1990s owing to their widespread detection in all environmental matrices on a global scale.⁶⁷⁻⁷¹ PBDEs are similar to polychlorinated biphenyl ethers (PCBs) in terms of their environmental behaviors and adverse impacts on human and ecological health.^{72, 73} The production and use of PBDEs have been banned in Europe since 2004. The import and production of PBDEs by the U.S. were voluntarily phased out at the end of 2004, and their use in many states in the U.S. has been banned. Virginia HB 2394 (2019; which did not proceed out of committee) proposed to prohibit, starting July 1, 2020, the manufacture or sale in the Commonwealth of upholstered furniture intended for residential use or any product that is intended to come into close contact with a person younger than 12 years of age if such product contains any flame-retardant chemical listed in the bill.⁷⁴ However, many products manufactured with PBDEs remain in use.

With the ban and phase out of PBDEs, there has been a rapid increase of production and usage of “alternative” or “novel” brominated flame retardants, e.g., decabromodiphenyl ethane (DBDPE), 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), hexabromobenzene (HBB), hexabromocyclododecane (HBCD), and pentabromoethylbenzene (PBEB) or other formulations, including organophosphate-based flame retardants. Some of the alternatives are known to be mutagenic and/or suspected to be neurotoxic.⁷⁵ For others, their impact on human and environmental health has been revealed only recently or remains to be further assessed.⁷⁶⁻⁷⁸

Flame retardants may be released into the environment from the production, use, and eventual disposal of products that contain them.^{79, 80} Discharges from manufacturing industries, wastewater treatment plants,⁸¹ as well as leachates from landfill sites are important point sources of these compounds.⁸²⁻⁸⁴ Important nonpoint sources include atmospheric deposition of emitted flame retardants with high volatility (especially PBDEs⁸⁵), processing/recycling and indiscriminate disposal of electronics and other wastes containing flame retardants,⁸⁶⁻⁸⁸ and biosolids/WWTP effluent applied to land.^{89, 90}

The Computer Recovery and Recycling Act established in 2008 in Virginia requires a computer manufacturer that makes or sells more than 500 units in Virginia to implement a recovery/recycling plan.⁹¹ About 20 Virginia localities offer ongoing collections of electronics or have conducted one-time electronics recycling collections, however, it is unclear where and how these collected electronic wastes are processed/recycled. This lack of information adds to the difficulty of assessing if electronic wastes are a possible source of flame retardants in the environment of Virginia. In 2017, 21.6 million tons of solid waste, about 60% of which was municipal solid waste and included unknown amounts of electronic waste, were received at

permitted solid waste management facilities in Virginia. Close to 76% of the solid waste managed in Virginia was landfilled on-site at the 51 permitted landfill facilities throughout the state.⁹²

The extent to which flame retardants leach out of landfills and into the waters of Virginia via direct discharge remains unknown. In Virginia, landfill leachate is often hauled to a WWTP for treatment or piped there directly. Landfills with direct discharge of leachate have individual permits. The DEQ data on permitted landfills were not reviewed for this report. These data should be reviewed for additional information on flame retardants (and other CECs). Effluents and biosolids discharged from WWTPs or septic systems are other likely sources of flame retardants in Virginia.

3.2. Occurrence

3.2.1. Aquatic Environment (Water, Sediment, and Organisms)

Many flame retardants are lipophilic (combine with or dissolve in lipids or fats) and have been shown to persist and accumulate in the environment. Because of their high hydrophobicity (repulsion from water) and high capacity to bioaccumulate (become concentrated within the bodies of organisms), worldwide environmental monitoring for flame retardants has focused on the occurrence of PBDEs in animal tissue, especially aquatic organisms, and in sediments in aquatic environments. Although there are no reports of PBDEs in the waters of Virginia, the reported levels in water elsewhere range from parts per quadrillion (ppq) to ppt.^{12, 54, 93} The levels of PBDEs reported in sediment and aquatic organisms from Virginia (Figure 3) are comparable with those reported elsewhere in the U.S. and worldwide.^{13, 14, 67} Chen and coworkers⁹⁴ reported that fishes collected from the Hyco River, Roanoke River, and Dan River in 2006–2007 generally exhibited HBCD concentrations that were orders of magnitude higher than those in 1999–2002. In contrast, the average fish PBDE concentration from all three rivers was about 4.5 times lower in 2006–2007 than in 1999–2002. These trends suggest HBCD use may have risen following the phase out and ban of PBDEs in consumer products. Currently, the occurrence of many “alternative” or “novel” flame retardants in the waters of Virginia is unknown.

3.2.2. Soil

Because of the high hydrophobicity and persistence of flame retardants, conventional WWTPs are not effective in degrading these compounds during wastewater treatment processes. The partition of PBDEs in biosolids has an influent-to-biosolids transfer coefficient in the range of 3–26 liters per gram (L/g), depending upon a given compound at a given WWTP.⁹⁵ An investigation⁹⁶ considered 77 traditional and novel brominated flame retardants in 110 biosolids composites collected from 97 U.S. WWTPs by the EPA for its 2001 national sewage sludge survey.⁹⁷ The reported average level of total PBDEs in the biosolids was 9.4 ppm (d.w.). This level decreased by about 50% in the biosolids surveyed by EPA just a few years later in 2006–2007.⁹⁸ This decrease is reflective of efforts to phase out PBDEs in the U.S. since 2004. An investigation of biosolids collected bimonthly during 2005–2011 from a mid-Atlantic WWTP serving 2.2 million people and generating 330 dry tons of biosolids/day reported total PBDEs concentrations of 563 ppb to 2.9 ppm (d.w.),⁹⁹ which were lower than those from the 2006–2007 EPA study. Since 2011, there have been no investigations of the occurrence of flame retardants, including many “alternative” or “novel” flame retardants, in biosolids from mid-Atlantic states.

Land applications of biosolids could be a potential route for flame retardants to enter aquatic environments. A 2009 survey examined PBDE data from 26 fields at 10 Virginia farms that received surface application of biosolids either multiple times (for up to 16 years) or a single application within 4 years. Three PBDEs (47, 99, 209) were found in the fields treated multiple times with biosolids at 110 ppb, and these PBDEs were found in the fields treated once within 4 years at 22 ppb.¹⁰⁰ The biosolids for this study were from a large mid-Atlantic WWTP and were applied to fields at rates that ranged from 42–54 dry tons per hectare (ton/ha). The total concentration of the three PBDEs tested in 2009 at these farms was 1.87 ppm. The median residence times were estimated to be 704 days for PBDE-47+PBDE-99 and 1,440 days for PBDE-209. With time, PBDEs in biosolids applied on land could potentially move into the surrounding aquatic environment via surface runoff. However, additional field studies are needed to confirm and quantify this impact.

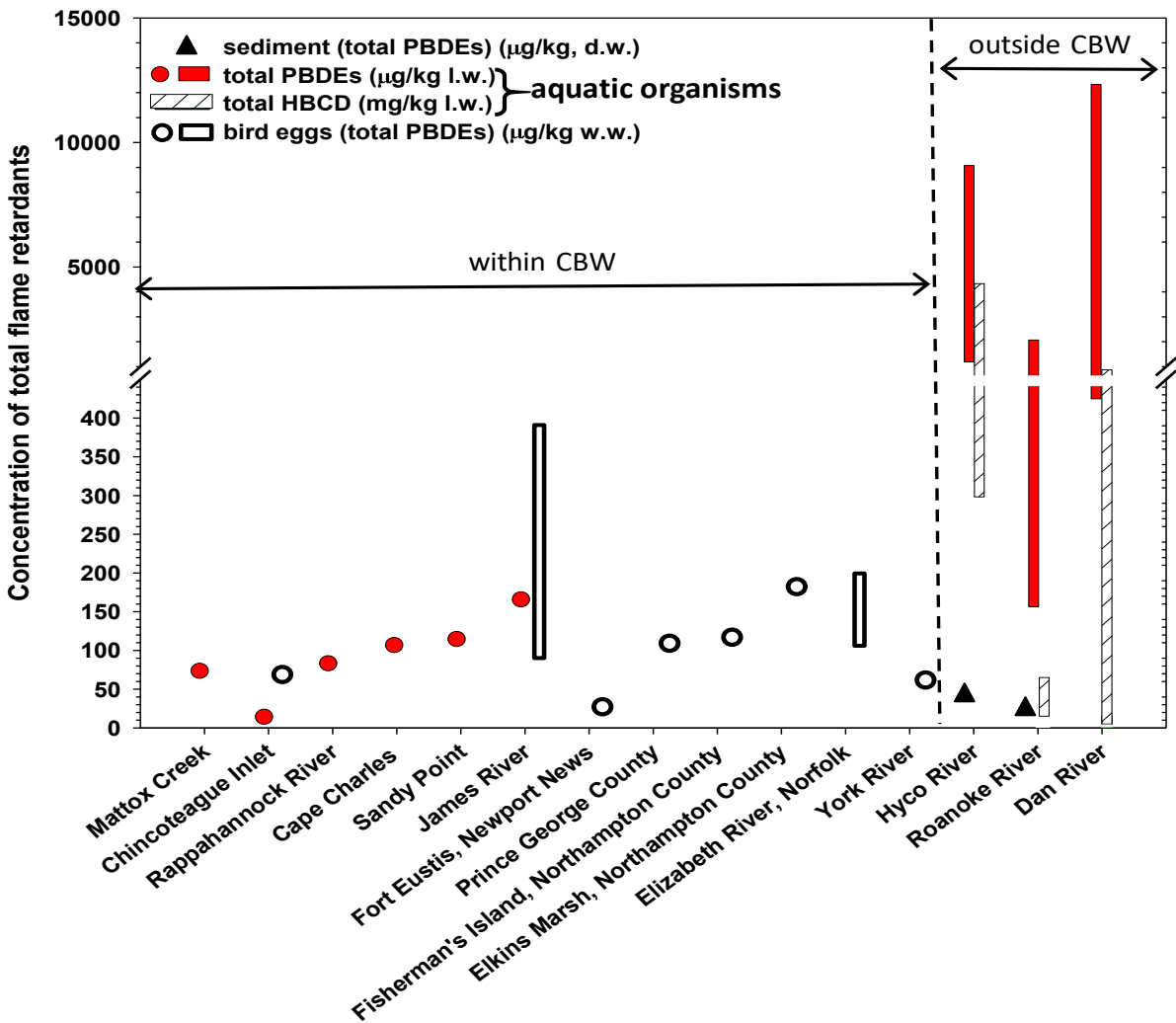


Figure 3. Flame retardants in the waters of Virginia at different locations. The circle represents the average concentration of a site. The bar represents the concentration range of a site. Table A2 in the Appendix includes detailed information pertaining to this figure. CBW = Chesapeake Bay watershed; $\mu\text{g}/\text{kg}$ = parts per billion (ppb); mg/kg = parts per million (ppm); d.w. = dry weight; l.w. = lipid weight; w.w. = wet weight

4. Hormones and Endocrine Disruptors

4.1. Major Sources

Natural and synthetic hormones, many PPCPs, and flame retardants are known or suspected to interfere with the functioning of endocrine receptors. The focus of this section is on hormonal compounds, which have been frequently detected and are proven potent endocrine disruptors in the environment.¹⁰¹ Natural hormones in vertebrates are synthesized by enzymatic modification of cholesterol and play a variety of roles in maintaining biological functions.¹⁰² They are classified as androgens, estrogens, or progestogens. Androgens are found at higher levels in males, and estrogens are found at higher levels in females. Progestogens are largely responsible for initiation and maintenance of pregnancy. The synthetic hormones ethynylestradiol (EE2) and mestranol (MeEE2) are used as contraceptives for humans,¹⁰³ and trenbolone acetate and zeranol are used for growth promotion in livestock.¹⁰⁴ Both natural and synthetic hormones are excreted in the urine and feces of both sexes of humans, pets, and farm animals, either in the free form or as glucuronide or sulfate conjugates. Depending on the species, the life stage of the individual, and other factors, concentrations of natural hormones in excreta can vary significantly, from non-detectable to ppm on a dry-weight basis.¹⁰⁵⁻¹⁰⁷

Natural hormones are ubiquitous in the environment because they are components of the life cycle of all organisms. Excreta from cats and dogs generally contains natural hormones (synthetic hormone use is much lower in pets than in humans and in livestock), and pet excreta typically is disposed of in landfills, is buried in soil, or is deposited on the ground. Natural and synthetic hormones excreted by livestock are concentrated in manure, and those excreted by humans are treated at WWTPs or septic systems and may eventually be concentrated in biosolids. In the U.S. more than 1 billion dry tons of animal manure are generated yearly,¹⁰⁸ and 13.8 million (0.0138 billion) dry tons of biosolids are produced yearly.¹⁰⁹ Each year, the CBW states produce approximately 44 and 1.3 million dry tons of animal manure and biosolids, respectively.¹¹⁰ In the U.S. the majority of the more than two billion farm animals are raised in CAFOs,¹¹¹ which produce large quantities of animal manure that are stored or applied on land as fertilizer. It is obvious that animal manure deposited on land and biosolids applied to agricultural fields could be possible major nonpoint sources of hormones. Because of the ineffectiveness of most conventional WWTPs for complete hormone degradation and removal, WWTP effluent could be a potential major point source of hormones.¹¹²⁻¹¹⁴

4.2. Occurrence

4.2.1. Aquatic Environment (Water, Sediment, and Organisms)

Although previous research has suggested that both CAFOs and WWTPs may be important sources of hormones in aquatic environments, little information is available on their occurrence in CAFO and WWTP effluents in Virginia (Figure 4). Elsewhere in the U.S., hormones have been detected in animal manure lagoons and effluents in the range of ppt to ppb¹²⁰ and in WWTP effluents at ppt levels.^{121, 122} An investigation of 18 stream sites in the Shenandoah River watershed demonstrated strong positive correlation ($R^2=0.39-0.75$) between watershed densities of CAFOs and in-stream concentrations of 17β -estradiol equivalents (E2Eq).¹¹⁵ (An E2Eq value can be determined with an estrogen-screening test. The test uses a bioluminescent yeast that reflects the estrogenicity of a water sample by exposing all estrogenic compounds in the water

sample.) A recent 2019 USGS survey of 21 hormones in the groundwater of 60 sites collected from Virginia reported no detection of hormones.⁴⁵

Environmental exposures of natural and synthetic hormones have been linked to intersex fish.^{116, 117} Kolpin and coworkers¹¹⁸ analyzed bed sediment at smallmouth bass nest sites in the Potomac River watershed and found a positive correlation between the total biogenic estrogens (17 β -estradiol, estrone, sistosterol, stigmastanol, and progesterone) in the sediments and the incidence of intersex fish using those nests. A later follow-up study on smallmouth and largemouth bass from 19 northeast national wildlife refuges in the U.S. found similar widespread occurrences of intersex fishes.¹¹⁹

A recent investigation of water, sediment, oysters, and mussels from 14 sites at Chester River, Manokin River, Holland Straits, and Kitts Creek along the Lower Eastern Shore of the Chesapeake Bay in Maryland detected estrone in all water samples at 500 ppq to 2.3 ppt but no detection of other hormones. Estrone in the sediment at eight sites ranged from 1.2–58 ppb (d.w.), and 17 β -estradiol was found at 11.5 ppb (d.w.) in the sediment of the site with the highest level of estrone.⁴⁶ This study also detected estrone, 17 β -estradiol, and 17 α -ethinylestradiol in mussels at 70, 16, and 15 ppb (d.w.), respectively. Furthermore, 17 α -ethinylestradiol was found in oysters at 19 ppb (d.w.) from one site on the Chester River. Information on the occurrence of hormones and endocrine disruption of aquatic organisms in waters outside the CBW is lacking for Virginia.

4.2.2 Soil

Hormones can enter the soil environment via land application of hormone-containing manure or biosolids¹²³ and result in their accumulation in soils up to ppb levels.¹²⁴⁻¹²⁶ A 2017 study of an agricultural field in Montgomery County, Va. evaluated hormone levels in soil 10 days after applying dry stack manure and dairy slurry that contained 11 hormones at tens to several hundred ppb. The study reported total hormone concentrations ranging from 0.39–7.38 ppb in surface soil (0–5 cm deep) and 0.43–1.74 ppb in deeper soil (5–20 cm deep). The level of progesterone in this field persisted until almost 200 days after manure application.¹²⁵ A similar trend was observed for an agricultural field in the Shenandoah Valley of Virginia where poultry litter had been applied.¹²⁵

It is generally thought that surface runoff and leaching may transport hormones from fields applied with manure or biosolids into the aquatic environment.¹²⁷⁻¹³² For example, a survey of 50 surface waters sampled in Delaware after manure applications in mid-spring detected estrone at 0–4 ppt, estrone-3-sulfate at 0–5 ppt, and 17 β -estradiol at 0–6 ppt.¹³³ These findings suggest runoff from the fields transported hormones to the surface water. However, because of the high hydrophobicity of hormones, their runoff and leaching potential is likely to be dependent on the type of soil (the octanol-water partition coefficient, log K_{ow} , of hormones ranges from 2.6 to 4.0). A recent rainfall simulation study in Montgomery County, Va. demonstrated that manure-borne hormones were not transported in runoff from a field with silty or loamy soils that had been applied with liquid dairy manure.¹²⁵ In addition, the method of land application may significantly affect runoff from fields. In the Montgomery County study, subsurface injections of manure further decreased the likelihood of hormone export off the field via surface runoff compared to surface applications.¹²⁵

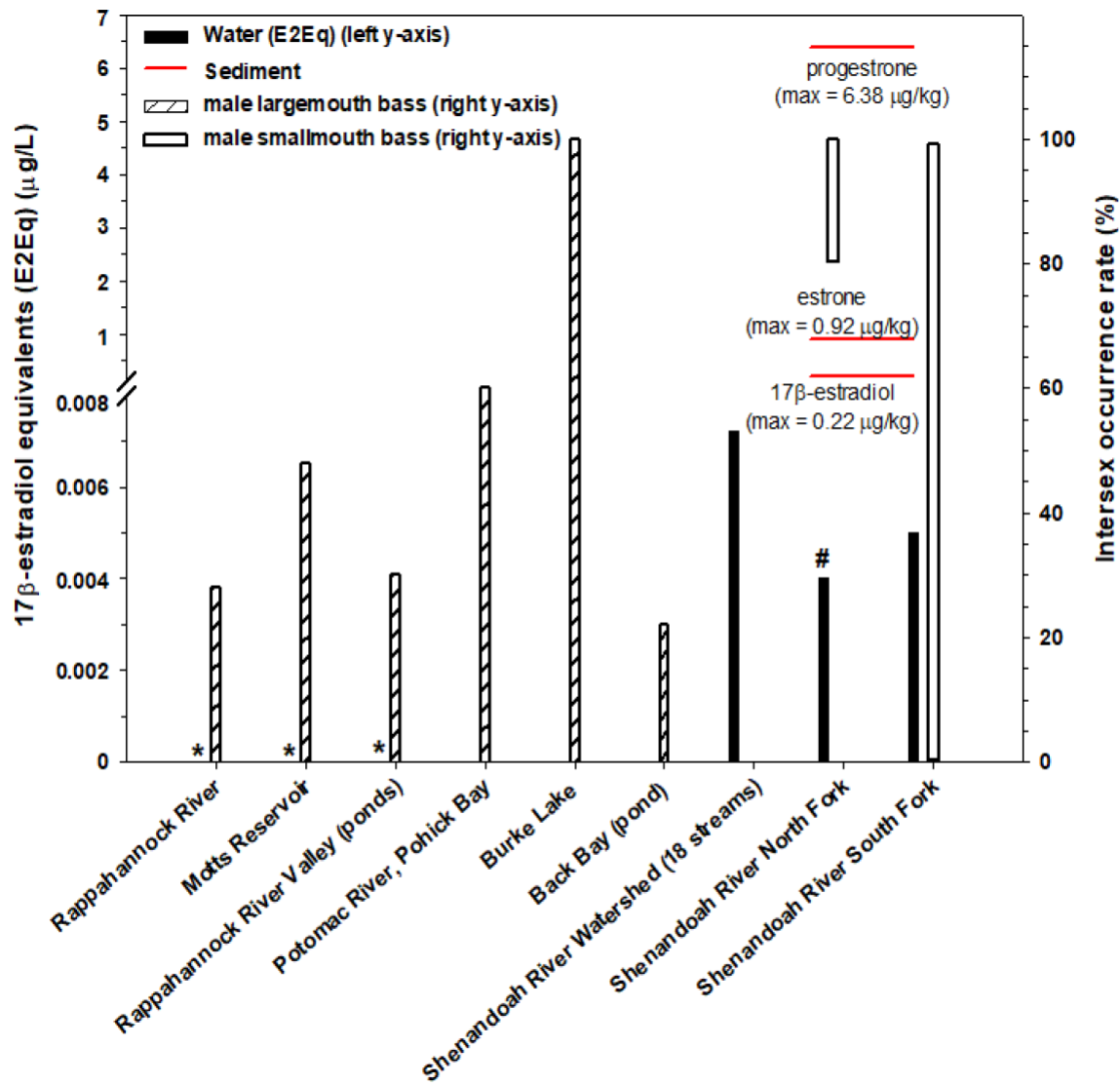


Figure 4. Maximum levels of estrogenicity as expressed in E2Eq in water, hormones in sediment, and the occurrence of intersex fishes in different Virginia waters in the Chesapeake Bay watershed. Table A3 of the Appendix provides detailed information pertaining to this figure.

* indicates detectable E2Eq.

indicates that the following compounds were detectable in the water: 17α-ethynylestradiol, 17α-estradiol, 17β-estradiol, androstenedione, and estrone.

μg/L and μg/kg = parts per billion (ppb)

5. Perfluorinated Compounds

5.1. Major Sources

Perfluorinated compounds (PFCs) are a broad range of compounds used in numerous applications, including stain repellents for textiles, additives to paper products, and materials in aqueous film-forming foams (AFFFs) used to control electrical fires. Per- and polyfluoroalkyl substances (PFASs, $C_nF_{(2n+1)-R}$) are man-made fluorinated aliphatic (carbon chain) compounds, which are a subgroup of PFCs that includes any organic compound that contains fluorine.^{134, 135} Since the 1940s, more than 3,000 PFASs have been produced and used in a wide range of industrial and consumer applications.

Of the long list of PFASs (Figure 5), the following have attracted worldwide regulatory attention since the 2000s because of their persistence and human health concerns:

- Perfluoroalkyl carboxylic acids (PFCAs) with seven or more perfluorinated carbons;
- Perfluoroalkane sulfonic acids (PFASs) with six or more perfluorinated carbons; and
- Individual compounds of PFCAs and PFASs, in particular perfluorooctanoic acid (PFOA) and perfluorooctanesulfonate (PFOS).¹³⁵⁻¹³⁹

Although production of most long-chain PFASs (more than six carbons) has been phased out in the U.S. and has been generally replaced by production of shorter chain PFASs, they are still produced and used elsewhere.

Both PFOA and PFOS were listed on the 2016 EPA Contaminant Candidate List (CCL 4) for public drinking water systems.²⁷ The Third EPA Unregulated Contaminant Monitoring Rule (UCMR 3) included PFOA and PFOS, as well as perfluoroheptanoic acid (PFHpA), perfluorononanoic acid (PFNA), perfluorobutane sulfonic acid (PFBS), and perfluorohexane sulfonic acid (PFHxS). In 2016, the EPA established a lifetime health advisory level in drinking water of 70 ppt for PFOA and PFOS, individually, or in combination.^{140, 141} Currently, 19 states in the U.S. (but not Virginia) have established advisory standards for PFASs in drinking water. Some of the state advisory standards are stricter (as low as 10 ppt) compared to the EPA advisory levels and include other PFASs (up to 15 compounds) in addition to PFOA and PFOS. Other state advisory limits are less strict (up to 300 ppt) compared to the EPA advisory and are focused only on PFOA and PFOS.¹⁴²

Regulations on PFASs and evidence of their adverse human and environmental health impacts are rapidly evolving.^{18, 138, 143, 144} In early 2019, EPA released a PFAS Action Plan that includes the development of maximum contaminant levels (MCL) for PFOA and PFOS and offers to assist states, tribes, and communities in addressing PFASs.¹⁴⁵ Comprehensive background information on PFASs and ongoing research by EPA and the Centers for Disease Control and Prevention (CDC)/Agency for Toxic Substances and Disease Register (ATSDR) can be found elsewhere.^{142, 146, 147}

Replacement chemicals for some PFASs have recently drawn increasing attention owing to their wide usage, detection in waters, and various suspected human health impacts.^{148, 149} GenX chemicals, which replace PFOAs, include hexafluoropropylene oxide (HFPO) dimer acid and its ammonium salt. Likewise, perfluorobutane sulfonic acid (PFBS) is a replacement for PFOS. In

early 2019, the EPA released draft toxicity assessments for both of these chemicals, HFPO¹⁵⁰ and PFBS.¹⁵¹

Although PFASs are used in a variety of consumer products,^{152, 153} the major human exposure to PFASs is believed to be drinking water contaminated with PFASs.¹⁵⁴ It has been estimated that drinking water supplies for 6 million U.S. residents exceed the lifetime health advisory (70 ppt) for PFOA and PFOS established by EPA.¹⁷ The major sources of PFASs in drinking water are assumed to be localized and associated with industrial facilities where PFASs were/are produced or used to manufacture other products. Other sources include oil refineries (none in Virginia), airfields, closed and active military installations, or other locations where PFAS were used for firefighting with AFFF.^{17, 155-158} The 2019 record by the Federal Aviation Administration (FAA) lists 81 active airfields in Virginia, 11 of which are for military use.¹⁵⁹ There are a total of 26 military installations in Virginia, several of which have identified potential contamination of PFASs in nearby groundwater.^{160, 161} There are 16 firefighting training facilities in Virginia. To date, the focus has been on facilities where PFASs-containing products have been directly manufactured or used.

Investigations have suggested that septic systems,¹⁶² WWTP effluents,¹⁶³ biosolids,¹⁶⁴⁻¹⁶⁶ and legacy or current landfills^{144, 167, 168} are other important sources of PFASs in the environment. Furthermore, atmospheric deposition has recently been proposed as a significant source of PFASs.¹⁶⁹⁻¹⁷³ Once released to the environment, PFASs are highly resistant to physical, chemical, and biological degradation and are persistent in the environment because of the strong structural carbon-fluorine bonds they contain.^{174, 175} They are also bioaccumulative because of their high K_{ow} values.¹⁷⁶

5.2. Occurrence

A recent literature review¹⁷⁶ summarizing PFASs-related publications between 2009 and 2017 has identified 455 PFAS compounds, of which 45% are anionic (negatively charged), 29% are zwitterionic (containing separate positively and negatively charged groups), 17% are cationic (positively charged), and 8% are neutral compounds. Some of the identified PFASs are potential precursor compounds of PFOA and PFOS. Throughout the world, various PFASs have been detected in drinking water,¹⁷ natural waters,^{15, 16} aquatic organisms,^{177, 178} sediments,^{16, 179} wastewater,¹⁶³ biosolids,¹⁶⁴⁻¹⁶⁶ and soils.¹⁸⁰ However, there is limited information on their occurrence in the environment of Virginia.

5.2.1. Aquatic Environment (Water, Sediment, and Organisms)

A 2016 publication¹⁷ that summarizes the publicly available concentration data from the UCMR 3¹⁴⁰ reported detection of PFOA and PFOS in drinking water samples collected from water utilities in central and eastern Virginia. The concentrations in these samples were reported at 20–70 ppt for PFOA and at 40–200 ppt for PFOS. However, a close inspection of the original UCMR 3 data showed levels in all tested Virginia drinking water samples below the minimum reporting level (MRL) of 20 ppt for PFOA and 40 ppt for PFOS. The levels of other tested PFASs were also below the MRL, e.g., MRLs for other tested PFASs: PFNA=20 ppt, PFHxS=30 ppt, PFHpA=10 ppt, and PFBS=90 ppt.

As shown in Table A4 in the Appendix, a 2016 Navy investigation reported detection of PFOA and PFOS above the EPA lifetime health advisory level in monitoring wells and on-base drinking-water supply wells at the Naval Auxiliary Landing Field Fentress (Chesapeake, Va.) and in some off-site private wells. Similar monitoring results (Table A4) were reported for the on-site monitoring wells at the Naval Air Station Oceana (Virginia Beach, Va.) that is 12 miles north of the Naval Auxiliary Landing Field Fentress. The source of the PFAS at both sites is AFFF used for firefighting training activities conducted at the sites. The U.S. Navy is continuing to monitor the levels of PFASs in the groundwater and drinking water at those sites. The temporal and special changes of the levels of PFASs in the groundwater at the two sites are currently unknown. Although various PFASs were detected in 95 samples collected from 18 unnamed U.S. landfills at concentrations up to ppb,¹⁶⁸ it is unknown to what extent these compounds are present in the landfill leachate in Virginia.

Published concentrations of PFASs in surface water and sediment samples vary widely — over more than seven orders of magnitude. The reported median levels of PFASs in aquatic systems worldwide are up to several hundred ppt (d.w.) in sediments, whereas their levels are up to the low ppt in surface water; this finding suggests an overall tendency to accumulate in sediments.¹⁸¹ Compared to the reported levels of PFASs in the aquatic systems elsewhere in the world, aquatic systems in the U.S. have the highest average levels of PFASs.¹⁸¹ Variability in K_{ow} values among the PFASs would likely lead to a higher occurrence of certain isomers within different phases. For example, more linear and longer-chain isomers, which have a higher K_{ow} compared to those with more branched and shorter-chain isomers, are likely to occur more frequently in organic matter fractions of sediment, and shorter-chain, branched congeners are more likely to occur in the aqueous phase.¹⁸² Although PFASs have been detected in groundwater at two military installations in Virginia, information is lacking on their occurrence in surface waters and sediment in Virginia.

Recent assessments showed widespread detection of PFASs in various aquatic organisms, including PFASs with a wide range of chain lengths and configurations and their precursors. PFOS is the predominant PFAS found in all species, tissues, and locations analyzed worldwide (mean concentrations up to 1.9 ppm [wet weight, w.w.]), followed by long-chain PFCAs (Σ PFCAs up to 400 ppb [w.w.]).¹⁸³ The only record¹⁸⁴ of detection of PFASs in Virginia aquatic organisms was published in 2004. This study sampled osprey eggs from the Elizabeth River and the Middle Potomac River and reported detection of PFOA, PFOS, perfluorodecanoic acid (PFDA), and perfluorodecane sulfonic acid (PFDS) at up to several hundred ppb (w.w.) (Table A4 of the Appendix).

5.2.2. Soil

Because of their high K_{ow} values,¹⁷⁶ many PFASs and their precursors have high partition potential into the solid phase during wastewater treatment processes. This property results in their accumulation in biosolids at levels up to ppm (d.w.).^{164, 179} It was reported that PFOS was the dominant PFAS in typical municipal biosolids (80–219 ppb, d.w.) as well as in biosolids-applied soils (2–483 ppb, d.w.).¹⁶⁵ The reported concentrations of all PFASs in soil increased linearly as the loading rate of biosolids increased. This study also demonstrated that the leaching potential of a PFAS decreases with increasing chain length, suggesting higher transport potential

for short-chain PFASs in soils amended with biosolids.¹⁶⁵ Furthermore, a variety of precursors of PFASs in biosolids could be transformed to PFASs over time in soils applied with biosolids.

A PFAS study included 32 surface soil samples collected from locations lacking direct human activity in North America (26 samples were from 18 states in the U.S., but none were from Virginia). Quantifiable levels of perfluoroalkyl carboxylates (PFCAs: perfluorohexanoic acid [PFHxA]-perfluorotetradecanoic acid [PFTeDA]) were found in all samples; the total concentrations of PFCAs ranged from 29 ppt to 14.3 ppb (d.w.). Furthermore, quantifiable perfluoroalkane sulfonates (PFSAs: PFHxS, PFOS and PFDS) were found in all but one sample, and their concentrations ranged from below the limit of quantification (LOQ) to 3.27 ppb (d.w.).¹⁸⁰ This study reported that PFOA and PFOS were the most commonly detected analytes at concentrations up to 2.67 ppb for PFOA and 3.1 ppb for PFOS. It was suggested that the atmospheric long-range transport of neutral PFASs followed by their oxidation and deposition provided a significant source of PFCAs and PFSAs to soils. Again, the occurrence of PFASs in biosolids and the soil environment in Virginia is unknown.

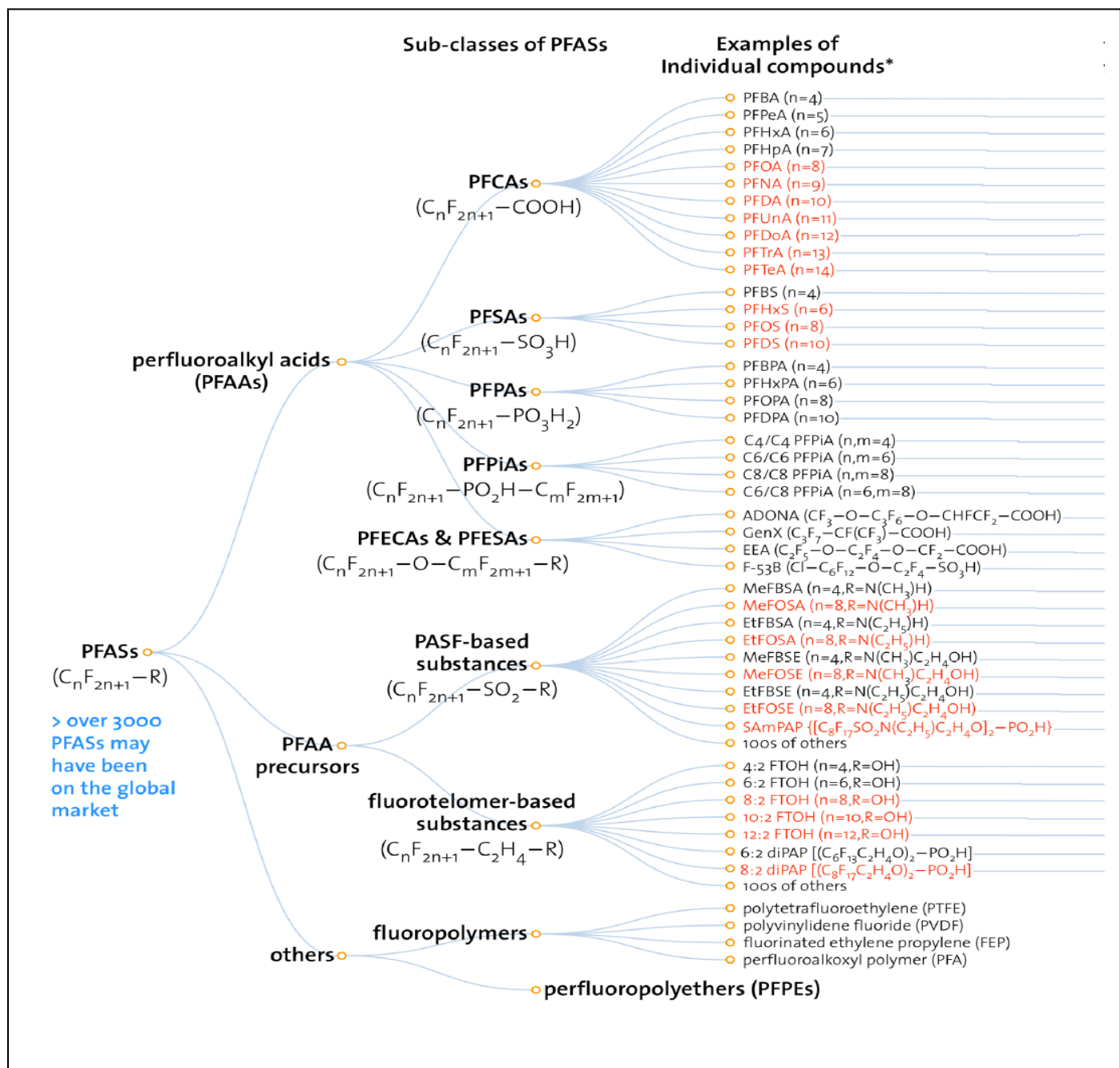


Figure 5. “Family tree” of PFAS, including examples of individual PFASs. Red text indicates PFASs that have been restricted under national/regional/international regulatory or voluntary frameworks with or without specific exemptions. Adapted with permission from Wang, Z.; DeWitt, J. C.; Higgins, C. P.; Cousins, I. T. A never-ending story of per- and polyfluoroalkyl substances (PFASs)? *Environmental Science & Technology* 2017, 51, (5), 2508-2518. Copyright 2017 American Chemical Society.

6. Antibiotics, Antibiotic-Resistant Microorganisms, and Antibiotic-Resistant Genes

6.1. Major Sources

About 80% of all antibiotics sold yearly in the U.S. are for therapeutic and sub-therapeutic uses¹⁸⁵⁻¹⁸⁷ in the production of food animals. Agriculture in the U.S. raises nearly 10.2 billion livestock annually; this livestock accounts for more than half of the U.S. agricultural cash receipts and exceeds \$100 billion/year in revenue.¹⁸⁸ In 2017, more than 12.3 million pounds of antibiotics important to human medicine and 24.1 million pounds of non-human antibiotics were approved by the U.S. Food and Drug Administration (FDA) for use in food-producing animals.¹⁸⁷

Up to 95% of antimicrobials administered to livestock can eventually be excreted through urine or feces as parent compounds or metabolites,^{108, 189} thereby resulting in their frequent detection at significant levels in manure.¹⁹⁰⁻¹⁹⁷ Similarly, antibiotics for human use are excreted and eventually enter into WWTPs and septic systems, which results in their frequent detection in wastewater, biosolids, and septic effluents.^{162, 198-202}

Antimicrobial-resistant elements (AREs) include antibiotics, antimicrobial-resistant microorganisms (ARMs), and antimicrobial-resistant genes (ARGs). Research has demonstrated that the overuse and/or misuse of antimicrobials can facilitate the development and proliferation of antimicrobial resistance in the microbiome of animal digestive systems;²⁰³ this process can result in elevated populations of ARMs and ARGs in animal manure, WWTP effluents, and biosolids.²⁰⁴⁻²⁰⁸ Thus, similar to hormones (Section 4.1), animal manure, WWTP effluents, and biosolids are major sources of AREs.^{108, 209-211} In addition, aquaculture is another major source of AREs in the environment.^{212, 213}

Widespread environmental occurrence and persistence of human and veterinary antibiotics may further stimulate the development of ARMs.²¹⁴ Overuse of antibiotics increases the likelihood that human pathogens can acquire antibiotic resistance through the transfer of resistant genes from the environment.²¹⁵⁻²¹⁹ Consequently, drug-resistant microbes reduce the effectiveness of antibiotic therapy for infectious disease.^{220, 221} Antibiotic resistance that compromises the effectiveness of antibiotics can lead to longer hospital stays, higher medical costs, and increased mortality. Such findings prompted the World Health Organization to declare antibiotic resistance as one of the most critical global human health challenges of the 21st century.²²² Annually, antibiotic resistance is believed to cause 48,000 deaths and more than \$3.5 billion in costs for health care in the European Union and U.S.²²³⁻²²⁵ Furthermore, research has shown that the presence of antimicrobials in the aquatic environment, even at ppt levels, can negatively impact reproductive, developmental, and other biological processes in mammalian and non-mammalian species.^{226, 227}

6.2. Occurrence

Although some antibiotics are naturally occurring,²²⁸ antibiotics have been detected at elevated levels of up to tens or hundreds of ppt or ppb in aquatic systems and soils.^{12, 229-231} Similarly, ARMs and ARGs have been detected in various environmental matrices.²³²⁻²³⁴

6.2.1. Aquatic Environment (Water, Sediment, and Organisms)

As shown in Figure 6, a wide range of concentrations of several antibiotics has been reported in surface water, groundwater, and drinking water in Virginia. A recent investigation of water from the Chester River, Manokin River, Holland Straits, and Kitts Creek areas along the Lower Eastern Shore of the Chesapeake Bay in Maryland reported seven commonly used antibiotics (azithromycin, clarithromycin, difloxacin, enrofloxacin, norfloxacin, roxithromycin, and sulfamethoxazole) at levels of 600 ppq to 114 ppt.⁴⁶ A recent 2019 USGS survey of groundwater from 60 sites collected from Virginia reported no detection of five tested antibiotic compounds including erythromycin, sulfadimethoxine, sulfamethizole, sulfamethoxazole, and trimethoprim.⁴⁵ However, this survey reported that at a national level, sulfamethoxazole was detected at 19–120 ppq in more than 0.5% (more than 5) of the groundwater samples from the 1,091 sites across the U.S.

Although frequent detection of antibiotics in sediments has been reported for various aquatic systems elsewhere,^{12, 229, 235} no information can be found for sediments in Virginia. Oxytetracycline was detected at 0.6–3.3 ppb (d.w.) in the sediments from two waterbodies (Yellow Bank Stream and Pocomoke River) on the eastern shore of the Chesapeake Bay in Maryland.²³⁶ As reported in the literature, sediment samples in general showed a greater detection frequency and a much higher concentration for antibiotics (ppb levels) compared to aqueous samples (ppt levels) taken at the same site (concentrations of antibiotics were 1,000 times higher in sediment samples than in water samples). This finding is likely a result of the sediment/water partition coefficients for antibiotics being at several thousand liters per kilogram (L/kg), a factor that is positively correlated to the hydrophobicity or organic carbon-water partition coefficient ($\log K_{oc}$) of the compound.^{228, 237}

It has been hypothesized that heavy use of antibiotics and synthetic antimicrobial agents may contribute to the selection pressure for microorganisms in the aquatic environment.²³⁸ Resistance selection concentrations recently predicted for 111 antibiotics ranged from 8 ppt to 64 ppb.²³⁹ Strong positive correlations between concentrations of antibiotics and levels of ARGs have been found in the sediments of aquatic systems heavily impacted by human activities.^{240, 241} This finding suggests that sediments can act as a reservoir of ARMs and ARGs.²³²

Although levels of antibiotics in aquatic biota have rarely been reported in the waters of Virginia or within the CBW, their bioaccumulation in aquatic organisms has been reported elsewhere at concentrations up to ppb (d.w.) levels. Bioaccumulation by organisms includes the uptake of the substance of interest from water as well as from the diet of the organism. There are several ways to measure and assess bioaccumulation in organisms, one of which is the bioaccumulation factor (BAF). The EPA uses different methods to estimate the BAF of organic chemicals, including antibiotics.²⁴³ BAFs can be calculated using either empirical data or measurements as well as from mathematical models. Antibiotics exhibit a wide range of BAFs, from tens to several hundred thousand L/kg.^{231, 237, 242} It is well known that, similar to other organic compounds, the bioaccumulation of antibiotics by aquatic organisms is affected by different factors, e.g., the lipophilicity/hydrophobicity of the antibiotic as indicated by its octanol-water partition coefficient (K_{ow}) and its degree of ionization as indicated by its acid dissociation constant (pK_a). The pK_a is affected by the pH of the water. Antibiotics that are in

neutral form at typical pH levels for natural water and are poorly water soluble ($\log K_{ow} > 1$) tend to accumulate in the fatty tissues of organisms.²²⁹

Elevated levels of antibiotic-resistant bacteria in the water and shellfish of the Chesapeake Bay were reported as early as 1976.²⁴⁴ This study showed higher numbers of antibiotic-resistant coliform bacteria near populated areas such as Baltimore. The authors hypothesized that WWTP effluents had a detectable influence on the water quality of the Chesapeake Bay. Their work showed that of 34 isolates from water and shellfish samples, many exhibited resistance to antibiotics, e.g., 58.8% to tetracycline, 58.8% to streptomycin, 32.4% to kanamycin, 32.4% to sulfadiazine, 11.8% to chloramphenicol, and 5.9% to gentamicin. Furthermore, many exhibited resistance to multiple antibiotics, e.g., 25.9% to ampicillin and tetracycline, 73.7% to ampicillin and streptomycin, and 58.8% to three or more antibiotics.

In 2014, surface water samples from Sandy Point State Park and the St. Martin River, which are two frequently used recreational areas in the Chesapeake Bay, and Pocomoke Sound, which is a heavily used commercial fishing area in the Chesapeake Bay, were evaluated for antimicrobial resistance among *Vibrio vulnificus* and *V. parahaemolyticus*.²⁴⁵ Both microbial strains can cause *Vibrio* infections originating from dermal contact with waters laden with *Vibrio* or through ingestion of seafood originating from such waters. This study found that 78–96% of isolates expressed intermediate resistance to chloramphenicol and 68% of *V. parahaemolyticus* isolates were resistance to penicillin.

Because a number of *Mycobacterium cosmeticum* isolates were recovered from otherwise healthy patients with infections acquired from direct exposure with the waters of the Chesapeake Bay,²⁴⁶ a follow-up study was conducted. This study isolated *M. cosmeticum* from surface waters of the Chesapeake Bay at Sandy Point State Park and reported its resistance to several antibiotics, including doxycycline, tigecycline, clarithromycin, trimethoprim/sulfamethoxazole, imipenem, cefoxitin, ethionamide, and streptomycin.²⁴⁷ *Mycobacterium cosmeticum* is commonly found in salt water and freshwater and is the causative agent of disease in many species of fish and occasionally in humans. This study raised important questions regarding the possible causes for the emergence of antibiotics resistance in this isolate:

- 1) Is there a source of environmental exposure to antibiotics?
- 2) Is it a human isolate transferred to the watershed?
- 3) Is it the result of lateral gene transfer with other resistant organisms in the Bay?
- 4) How do antibiotic use and disposal, not only in human medicine but also in agriculture, affect the development of antibiotic resistance in aquatic microorganisms that are significant to human health?

Although elevated ARE levels in aquatic systems have been frequently observed elsewhere and clear influences of human activities have been reported,^{231, 248} the occurrence of AREs in the waters of Virginia is unknown.

6.2.2. Soil

The overall occurrences of AREs in animal manure, biosolids, and soils receiving applications of wastewater have been summarized, and the reported levels of antibiotics are up to

several hundred ppb (d.w.).^{230 249} Elevated ARM populations and ARGs have been reported for soils receiving long-term applications of manure, biosolids, or wastewater.²⁵⁰⁻²⁵²

A recent study conducted on Southwest Virginia farmland showed surface runoff could be a significant route for transporting antibiotics from fields receiving applications of manure, amounting to 0.45–2.62% of their initial input with manure. Compared with surface applications of manure, subsurface injections of manure reduced the transport of the following antibiotics in runoff: sulfamerazine by 47%, chlortetracycline by 50%, pirlimycin by 57%, and tylosin by 88%.²⁵³ Furthermore, this study also demonstrated that manure application at least 3 days before a subsequent rain event could reduce antibiotic surface runoff by 9 to 45 times.

A follow-up investigation²⁵⁴ on the same Southwest Virginia fields demonstrated that 20–50% of the fecal coliforms associated with surface-applied manure that were resistant to sulfamethazine, tetracycline, or erythromycin could leave the field via runoff during the first rain event that occurred 1 day after manure application. Similar to what was observed for antibiotics in runoff, subsurface injection of manure could significantly reduce the runoff of antibiotic-resistant fecal coliforms by up to 322–830 times compared to surface application. The study also showed that prolonged time gaps between manure application and subsequent rain resulted in additional reductions of runoff. A concurrent investigation in the same area demonstrated that exposure to dairy manure from animals fed antibiotics could lead to greater antibiotic resistance in soil microbial communities.²⁵¹ It is unknown if similar observations would be made for soils elsewhere in Virginia.

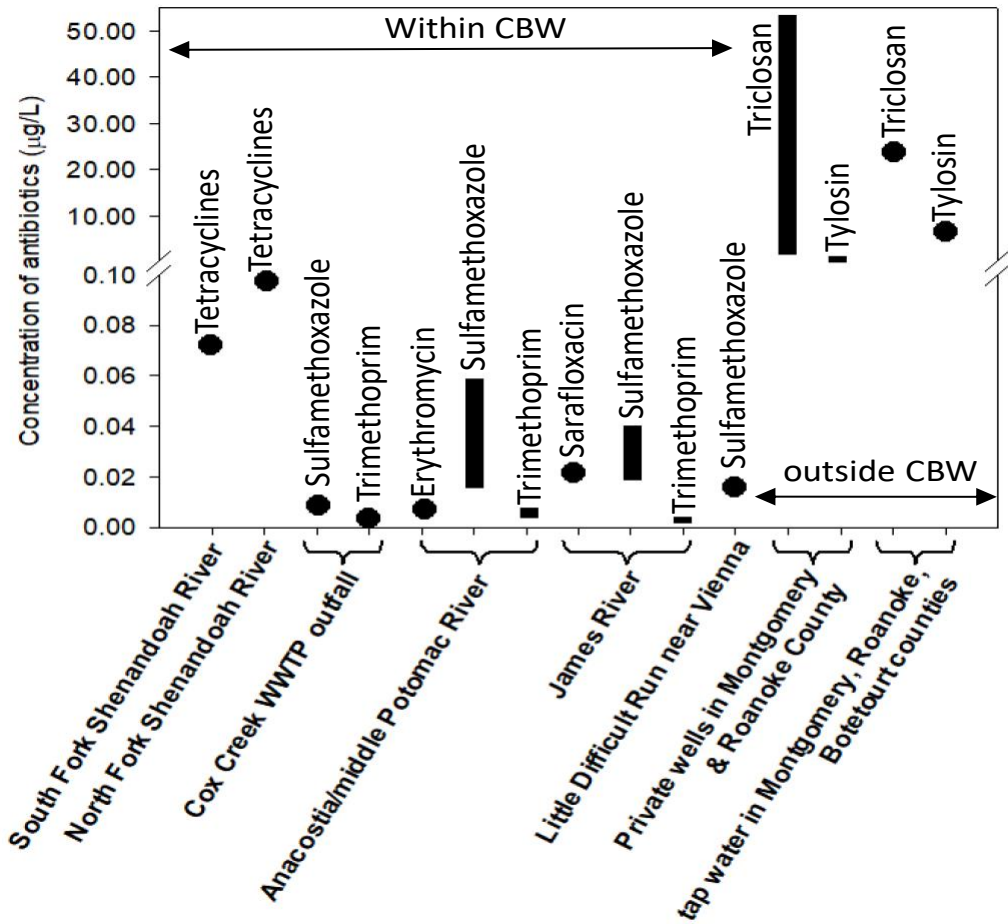


Figure 6. Concentrations of antibiotics in Virginia waters. Black dots indicate one concentration value, and a black bar shows the concentration range. Table A1 of the Appendix provides detailed information pertaining to this figure. CBW = Chesapeake Bay watershed; µg/L = parts per billion (ppb)

7. Micro- and Nano-Plastics and Microfibers

7.1. Major Sources

Since the 1950s, the production of plastics (polymer-based materials) has grown exponentially. Plastics have become one of the most-used anthropogenic products worldwide. In 2017, more than 348 million tons of synthetic polymers were produced and used worldwide in packaging, beverage bottles, and building and construction. Additionally, more than 70 million tons of synthetic polymers were produced and used in textiles, mostly for clothing and carpeting.²⁵⁵⁻²⁵⁷ Worldwide annual production of plastics is projected to rise to 1.8 billion tons by 2050, adding an extra 33 billion tons of plastics to the planet.²⁵⁸

Contamination by bulk plastics and plastic debris, including micro- and nano-plastics and microfibers, is currently one of the most widespread environmental problems in aquatic ecosystems worldwide.^{6, 19, 21, 259, 260} The term “microplastics/microfibers” commonly refers to synthetic particles with a dimension <5 millimeters (mm), which is estimated as the upper size limit for particles that can be readily ingested by aquatic organisms.²⁶¹ Nano-plastics include plastic debris with a dimension size <100 nanometers (nm); nano-plastics are of particular concern because they are more likely to pass biological membranes and affect cell function.²⁶² Another important environmental concern arises from the potential leaching of plastic constituents or sorbed contaminants after ingestion and subsequent biomagnification (the concentrating of toxins in an organism following its ingestion of food in which the toxins are more widely dispersed).²⁶³⁻²⁶⁷ Microfibers comprise more than 85% of microplastics found on shorelines around the world.²⁶⁸

Micro- and nano-plastics and microfibers are divided into two categories: 1) manufactured and deliberately used (either as resin pellets to produce larger items or directly in personal care products, textiles, and other products), and 2) formed from disintegration of larger plastic/fiber debris.²⁶⁹ A comprehensive review on the sources, occurrence, and environmental behavior of micro- and nano-plastics and microfibers in aquatic systems can be found in two open source books entitled *Freshwater Microplastics: Emerging Environmental Contaminants?*²¹ and *Marine Anthropogenic Litter*.²²

Domestic laundry discharges are the major source of microplastics and microfibers entering WWTPs; however, research has shown removal efficiencies for plastics at more than 96% from water during WWTP treatment processes, which results in WWTP effluent being an insignificant source of microplastics and microfibers compared to their overall environmental loads.²⁶⁸ Nonetheless, an estimated 8 trillion pieces of microplastics and microfibers enter the aquatic environment per day in the U.S. via WWTP effluents.²⁶⁹ Furthermore, up to 80% of the microplastics and microfibers entering WWTPs are eventually retained in biosolids, making biosolids a potential significant source.²⁷⁰ Some of the largest sources of micro- and nano-plastics and microfibers to the environment include the following: 1) soils in agricultural settings that use plastic mulching or receive application of biosolids or WWTP effluent, and 2) soils in urban areas that receive runoff from roads, parking lots and/or artificial turf. The amount of plastic in soil is possibly larger than what is in the marine environment.^{271, 272} The estimated total yearly input of 107,000–730,000 tons of microplastics and microfibers to European and North American farmlands exceeds their estimated total accumulated burden of 93,000–236,000 tons in

surface water in the global oceans.²⁷² Microfibers not directly associated with laundry effluent have been found to be ubiquitous and abundant in both indoor and outdoor environments, suggesting atmospheric deposition as another source.²⁰ Currently, limited information is available on the significance of septic systems as a source of microplastics and microfibers in the environment.

Overall, only 6–26% of plastics used worldwide is recycled. Approximately 21–42% is deposited in landfills, and 25–28% is directly released into the environment through a variety of pathways including littering, direct disposal, and abrasion or maintenance of outdoor plastic goods and coated surfaces.²⁶⁹ Although a large portion of plastics is deposited in landfills, limited information is available about the fate of plastics in landfills and the extent that micro- and nano-plastics and microfibers enter the environment via landfill leachate.

7.2. Occurrence

Worldwide occurrences of micro- and nano-plastics and microfibers have been documented in water, sediment, aquatic organisms of both freshwater and marine environments, and soil. However, data comparisons for levels of occurrence of plastics in different environmental matrices and in different locations is challenging. Analytical challenges include a lack of standardized protocols for sampling and analysis and no standard unit of measurement.²⁷³⁻²⁷⁸ In addition, because of the limitations of available sample-collection methods, most investigations have focused on particles >300 micrometers (μm) in size, resulting in a likely underestimation of plastics of smaller sizes.

7.2.1. Aquatic Environment (Water, Sediment, and Organisms)

In contrast to other CECs, an issue with quantifying micro- and nano-plastics/microfibers in water is the use of non-standard units of measurement, making it difficult to compare amounts among different sites.^{279, 255} The occurrences of micro- and nano-plastics and microfibers have been reported for waters worldwide. Rivers are estimated to transport 70–80% of plastics that eventually arrive in the ocean.²⁷⁹ An investigation detected microfibers throughout the length of the Hudson River at an average of 0.98 microfibers/L.²⁸⁰ The authors estimated that 34.4% of the drainage area to the Hudson River watershed contributes an average 300 million anthropogenic microfibers into the Atlantic Ocean per day.²⁸⁰ In the Great Lakes, downstream of the highly populated Detroit and Cleveland metropolitan areas, the concentrations of microplastics ranged from 280,947–466,305 particles/square kilometer (km^2).²⁸¹ The average abundance of microplastics in a highly urbanized river were reported to be 2.4 particles/cubic meter upstream of WWTPs and 5.7 particles/cubic meter downstream of WWTPs.²⁸² In the northeast Atlantic, microplastics were found in 94% of all surface samples with an average density of 2.46 particles/cubic meter.²⁸⁰ A recent study reported detection of microfibers at 0.86–15.2 particles/L in 16 out of 17 water samples collected from springs and wells from two karst aquifers in Illinois. The presence of microfibers was consistently associated with other parameters, including phosphate, chloride and triclosan, suggesting effluent from septic systems as a source.²⁸³

A 2011 sampling of water from four estuarine tributaries within the Chesapeake Bay in Maryland reported detection of microplastics in all but one of 60 samples. The concentrations for these samples ranged from 5,534 to 297,927 particles/square km^2 .²⁸⁴ This study also found

positive correlations between the concentrations of microplastics in the tested waters with the population density and the proportion of urban/suburban development within watersheds where the water samples were collected. Furthermore, this study detected peaks in microplastic concentrations at three of the four sample sites shortly after two closely occurring major storm events. This finding is likely attributable to substantial runoff of terrestrial debris as well as resuspension of plastics already in aqueous systems. Currently, no published data about plastics are available for waters in Virginia.

Micro- and nano-plastics and microfibers have been detected in sediments from around the world using various reporting concentration units, e.g., up to several hundred thousand particles/square meter, several thousand particles/kg, several thousand particles/cubic meter, and several thousand particles/L.²⁷⁴ Research has suggested that freshwater sediments can act as a sink for plastic pollutants²⁸⁵ and result in concentrations of plastics in the sediment being up to several hundred thousand times higher than in the water.²⁸⁶ Currently the Marine Debris Program of the National Oceanic and Atmospheric Administration (NOAA),²⁸⁷ which was established through the 2006 Marine Debris Act, has focused its effort on occurrence, fate, and impact of microplastics in the U.S. marine environment, including the Chesapeake Bay. However, little information is available on the occurrence of plastic pollutants in sediments of both marine and freshwater systems in Virginia.

Nano-plastics may enter the food web via algae and bacteria or be assimilated by filter-feeding organisms.²³ Because the size of microplastics and microfibers is similar to plankton and other small organic particles, aquatic organisms can ingest these plastics along with their natural food sources. This type of accidental ingestion is particularly common for non-selective feeders, which filter large quantities of water and sediment for organic nutrients. Microplastics and microfibers have been detected in the digestive tracts of organisms from different trophic levels for more than 690 marine species from different habitats, strongly suggesting the possibility of trophic transfer.^{23, 288} Again, it is difficult to compare data from different sites because of non-standard units of measurement. For example, a recent review reported concentrations in different organisms at 0.026–83 particles/organism, 0.2–1.2 particles/g (w.w.), and 34–83% detection rates.²³ Recent investigations of the marine environment of Virginia and the Chesapeake Bay²⁸⁹ have clearly demonstrated widespread occurrence of macroplastics in the region. A 2016 technical review²⁹⁰ suggested high possibilities of widespread occurrence of microplastics and microfibers in aquatic organisms of the Chesapeake Bay. However, mostly due to analytical limitations, there is a lack of systematic investigation of their occurrence in aquatic organisms in the waters of Virginia.

7.2.2. Soil

The sources of micro- and nano-plastics and microfibers to soil include the following: inputs from agricultural practices, the influence of runoff and deposition, and the fragmentation of larger plastic debris due to littering.²⁷¹ Of these sources, inputs from agricultural practices are likely to be the most significant.

Agricultural soils can be a major receiver of micro- and nano-plastics and microfibers because of their close association with biosolids, which are often applied to land as a soil amendment, and WWTP effluent, which is often used for irrigation. It has been estimated that

between 125 and 850 tons of microplastics/million inhabitants are added annually to European agricultural soils where ~50% of biosolids are applied on land.²⁷² Similar occurrences would be expected in the U.S. because ~70% of biosolids are applied on land. The maximum microplastic loading for U.S. farmland is calculated to be as high as 9 to 63 tons/hectare (t/ha).²⁹¹ It is estimated that annually 63,000–430,000 tons of microplastics would be deposited in European farmland, and 44,000–300,000 tons of microplastics would be deposited in North American farmland.²⁷² Considering about 46% of all Virginia biosolids are applied to approximately 43,000 acres of farmland and forestland in Virginia,³⁹ a substantial amount of micro- and nano-plastics and microfibers can potentially enter the affected soil environment.

Another source of microplastics in agricultural soils includes plasticulture, the use of plastic products in agriculture, e.g., utilizing plastics for mulch, raised beds, drip irrigation, soil fumigation, windbreaks, disease prevention, weed and pest management, packaging containers and sacks, pots, strings, ropes, etc. The practice of plasticulture has been growing in popularity with vegetable farmers in Virginia, resulting in 55–75% of a typical field being covered by impermeable plastic when using this practice.²⁹² Fragments of plastics are left behind after agricultural use and may accumulate within soils. They may also further fragment to micro- or nano-plastics.^{291, 293} To date, analytical limitations have hindered an accurate assessment of the levels of micro- and nano-plastics and microfibers in soil samples. Their levels in the soils of Virginia and their impact to the waters of Virginia are therefore unknown.

8. Engineered Nanomaterials

1.1 Major Sources

Nanomaterials are relatively small, typically ranging in size between 1–100 nm in at least one dimension. Nanomaterials include three major groups: natural, incidental, and engineered.²⁹⁴ Naturally occurring nanomaterials are ubiquitously present in the environment and include volcanic ash, ocean spray, magnetotactic bacteria, mineral composites, etc. Incidental nanoparticles are often by-products produced from industrial processes and household activities. Engineered nanomaterials (ENMs) have been hailed as the “next industrial revolution” because of their unique properties that can be used for specific functions. ENMs have been widely used in industry, medicine, consumer products, agriculture, and many other fields.²⁹⁵⁻²⁹⁷ Common ENMs include carbon nanotubes (CNTs), metal nanoparticles, and metal-oxide nanoparticles. ENMs with one dimension in the nanoscale are layers, such as thin films and quantum wells. Quantum well wires and nanotubes are materials that are nanoscale in two dimensions, and quantum dots are nanoscale in three dimensions.^{297, 298}

Because of the rapid development and wide application of ENMs, the global nanotechnology market is projected to exceed \$125 billion by 2024.²⁹⁹ The chances for the emission of ENMs to the environment are therefore growing and have led to increased concerns in recent years for their potential risk and toxicity to human and ecological health.^{300, 301} The sources of ENMs to the environment are believed to be intentional and unintentional releases to solid and liquid waste streams from households, manufacturing sites, waste treatment plants, unwanted spills, and emissions to the air.²⁹⁸ Possible major sources of ENMs include landfills as well as effluent and biosolids from WWTPs.³⁰²⁻³⁰⁶ It was estimated that 63–91% of 2010 global ENMs production were disposed of in landfills, with the balance released into soils (8–28%), water bodies (0.4–7%), and the atmosphere (0.1–1.5%).³⁰⁷ Because of their unique properties and application in various products, the market of ENMs in the Chesapeake Bay states, although unknown, is most likely mirroring the global market-growth trend. Information on the nanotechnology market in the Chesapeake Bay states would help better assess the potential input of ENMs to the region.

1.2 Occurrence

In contrast to other groups of emerging contaminants, the environmental occurrence and concentrations of ENMs are still largely unclear and limited. Because of the difficulties in developing analytical and assessment methods³⁰⁸ and the unknown emissions of ENMs to the environment, data are lacking.³⁰⁹ Many existing analytical methods have been developed for laboratory-controlled simple matrices; however, these methods are not capable of testing environmental samples, which often have complex matrices.^{307, 308, 310, 311} Furthermore, existing methods also fail to clearly distinguish between natural/incidental and engineered nanomaterials in complex environmental samples.³⁰⁹

Currently, the occurrences of ENMs in the environment are largely based on theoretical models and are therefore limited to predictive values. Occurrences for selective ENMs in U.S. surface waters and soils were based on the production and usage of ENMs and the probabilistic material-flow analysis from a life-cycle perspective of ENMs-containing products. For U.S. surface water, the predictive occurrence levels ranged from 1.0 ppq for carbon nanotubes to 140

ppt for gold nanoparticles. In U.S. soil, the predictive occurrence levels ranged from 8.3 ppq for silver nanoparticles to 5.99 ppb for gold nanoparticles.^{298, 312}

A model was used to predict the concentrations of ENMs and their reaction byproducts in the water column and sediment of the James River basin. The model used 20-year historic weather data and the estimated 2010 zinc oxide and silver ENMs loading scenarios in wastewater effluent and biosolids. The developed spatially-resolved environmental-fate model estimated the influence of daily variation in streamflow, sediment transport, and stream loads from point and nonpoint sources.³¹² This model predicted that less than 6% of ENMs-derived metals were retained in the river and sediments of the James River basin due to the spatial and temporal variability in sediment transport rates. Agricultural runoff from fields applied with biosolids accounted for 23% of the total metal loads from ENMs to streams. The highest ENMs-derived metal concentrations in the water and sediment were predicted to be at several hundred ppt. Beyond the above predicted occurrence data, investigative data have not been found for the occurrence of ENMs in the waters of Virginia.

9. Conclusions and Recommendations

As described in the AAC FY 2019 scope of work, this report summarizes the sources and occurrence of identified emerging contaminants in the waters of Virginia. Table 1 shows the various sources for the seven groups of CECs discussed in the report. Our current knowledge of their occurrence in the waters of Virginia is limited. The occurrence data on PPCPs, flame retardants, hormones and endocrine disruptors, and antibiotics presented in this report are mostly from investigations conducted in the CBW. For waters outside the CBW, the occurrence of these four classes of CECs is largely unknown in Virginia. Although detection of ARMs in the Chesapeake Bay was reported as early as 1976, follow-up monitoring on the occurrence of ARMs and ARGs in the region has been limited. Investigative work on the occurrence of perfluorinated compounds in the CBW has focused on a few military installments and firefighting facilities. Otherwise, there is limited information on their occurrence in the waters of Virginia. Although plastic-waste contamination is widespread in the Chesapeake Bay states, there has been only one published investigative work detailing the observation of microplastics and microfibers in the waters of this region (and it does not include Virginia). No published information on the occurrence of plastic pollutants has been found for the waters of Virginia. A lack of sensitive analytical methods has hindered our assessment and better understanding of the environmental occurrence of nano-plastics and engineered nanomaterials.

In general, there is a lack of occurrence information on all groups of CECs in the waters of Virginia. Better understanding and identification of major sources of all groups of CECs in Virginia would help predict their occurrence. Future work should prioritize the CECs to monitor in Virginia by identifying the risk of the contaminants to humans and aquatic life. This effort should be guided by the known major health and environmental concerns and current EPA recommendations/advisories listed in Table 1. Future work should also focus on how best to monitor the CECs of greatest risk in Virginia. This information would be of great value to DEQ in developing an effective and efficient monitoring strategy to assess CECs in the waters of Virginia.

Table 1. Summary - Occurrence and sources of major CEC groups, challenges, major concerns, and existing EPA recommendation/advisory.

CEC groups	Occurrence				Sources						Major challenges	Major concerns	Existing EPA recommendation /advisory
	water	sediment	organisms	soil	WWTP effluent	biosolids	animal manure	landfill	other*	Atmospheric deposition			
Pharmaceuticals and Personal Care Products (PPCPs)	X	X	X	X	X	X		X			<ul style="list-style-type: none"> • New compounds are emerging • Analytical methods • Toxicological assessment 	Exposure	No
Flame Retardants		X	X	X		X		X		X		Endocrine disruption	Yes
Hormones and Endocrine Disruptors		X	X	X	X	X	X					Endocrine disruption	Yes
Perfluorinated Compounds	X	X	X	X?		X?		X	X		<ul style="list-style-type: none"> • >3000 compounds within this group • Analytical methods and toxicological assessment are limited to a small subgroup of compounds 	Adverse human health impacts including liver damage, thyroid disease, decreased fertility, high cholesterol, obesity, hormone suppression, and cancer	Yes
Antibiotics, Antibiotic Resistant Microorganisms, and Antibiotic Resistant Genes	X	X	X	X	X	X	X	?				Antibiotic resistance in humans	No
Microplastics and Microfibers	X	X	X	X	X	X		?		X	<ul style="list-style-type: none"> • Analytical methods • Toxicological assessment 	Unknown	No
Nanomaterials	X?	X?	X?	X?	X?	X?		X?	?	?	<ul style="list-style-type: none"> • Analytical methods • Toxicological assessment 	Unknown	No

*includes military installation, airport, firefighting training facilities, manufactures of the products, etc.

X, indicates significance

X?, expected to be significant based on model prediction but currently there is a lack of data

?, currently unknown or lack of information

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Appendix

Table A1. Occurrence of PPCPs in the waters of Virginia [#]					
List of identifiable compounds or concentrations				Locations	Ref*
Compounds	Water (ppb)	Sediment (ppb)	Organisms (ppb)		
<i>para</i> -Cresol <i>N,N</i> -diethyltoluamide (DEET) Caffeine Codeine Carbamazepine Venlafaxine	identifiable			North Fork of the Shenandoah River	313
<i>Sarafloxacin</i> ^{&}	0.02			James River near Richmond	314
Carbamazepine	0.012			South Fork Shenandoah River near Lynnwood	
Caffeine	0.0005-0.038			<ul style="list-style-type: none"> • Appomattox River near confluence with James River • Occoquan Reservoir • James River near Richmond • South Fork Shenandoah River near Lynnwood 	
Cotinine	0.005-0.074				
1,7-Dimethyl-xanthine	0.01-0.04				
<i>Tetracyclines</i>	0.076			South Fork Shenandoah River	118
	0.85			North Fork Shenandoah River	
16 PPCPs	identifiable	identifiable		South Fork Shenandoah River	
26 PPCPs		identifiable		North Fork Shenandoah River	
Carbamazepine	0.0056-0.03			Back River	315
Cotinine	0.0052-0.02				
Dehydronifedipine	0.0018-0.0027				
Caffeine	0.016			Cox Creek WWTP outfall	
Carbamazepine	0.0067				
Codeine	0.0014				
Cotinine	0.0052				
Diltiazem	0.0018				
<i>Sulfamethoxazole</i>	0.011				
<i>Trimethoprim</i>	0.0014				
Carbamazepine	0.0013				
Cotinine	0.0024			Annapolis WWTP outfall	
Caffeine	0.005				
Cotinine	0.0026			Chesapeake-Elizabeth WWTP outfall	
Carbamazepine	0.0011				
Cotinine	0.0025			Nansemond WWTP outfall	
Cotinine	0.0014				
Fluoxetine	0.0026				
Acetaminophen	0.064			Virginia Initiative WWTP outfall	316
<i>Erythromycin</i>	0.0113				
<i>Sulfamethoxazole</i>	0.0176-0.0588				
<i>Trimethoprim</i>	0.0036-0.0069			Anacostia/middle Potomac River	

Diphenhydramine	0.0012-0.0129		Catfish: 0.42 Gizzard shad: 1.02 Carp: 0.38	<i>Plasma of different organisms were tested</i>		
Atenolol	0.0448		Gizzard shad: 6.82			
Diltiazem	0.0011-0.0016		Gizzard shad: 0.42 Carp: 0.33			
Diclofenac	0.0122					
Gemfibrozil	0.0036-0.0092					
Carbamazepine	0.0072-0.0505		Catfish: 2.23			
Sucralose	0.699-4.59		Gizzard shad: 2301			
Methylphenidate	0.00105					
Caffeine	0.0571-0.1					
<i>Sulfamethoxazole</i>	0.0217-0.0413			James River		
<i>Trimethoprim</i>	0.0024-0.0029					<i>Plasma of different organisms were tested</i>
Diphenhydramine	0.00037-0.0017		Catfish: 0.51 Gizzard shad: 0.39			
Atenolol	0.0121-0.0136					
Diltiazem	0.0030-0.0075		Catfish: 0.77 Gizzard shad: 0.57			
Gemfibrozil	0.0030-0.0061					
Carbamazepine	0.0123-0.0159					
Sucralose	1.717-2.813					
Caffeine	0.0402-0.0691		Catfish: 10.9			
Acetaminophen	0.0023-0.0129			Accotink Creek near Annandale	10	
Caffeine	0.0429-0.148					
Metformin	0.0127-0.0197					
Methyl-1H-benzotriazole	0.335-0.688					
Nicotine	0.0156-0.0904					
Acetaminophen	0.006-0.0139					Difficult Run above Fox Lake, near Fairfax
Caffeine	0.0238-0.149					
Carbamazepine	0.0023-0.0059					
Cotinine	0.0021-0.0244					
Metformin	0.008-0.0265					
Methyl-1H-benzotriazole	0.282-0.766					

Nicotine	0.0803-0.1554				
Acetaminophen	0.0103-0.0831			Difficult Run near Great Falls	
Caffeine	0.021-0.0543				
Carbamazepine	0.0045-0.0066				
Cotinine	0.0038-0.0076				
Metformin	0.0081-0.0683				
Methyl-1H-benzotriazole	0.117-0.404				
Nicotine	0.0084-0.0514				
Caffeine	0.0126-0.080			Little Difficult Run near Vienna	
Carbamazepine	0.0149-0.0284				
Sulfamethoxazole	0.0157				
Metformin	0.0048				
Methyl-1H-benzotriazole	0.0168				
Nicotine	0.0549				
142 PPCPs were screened	73 were identified in at least one well			33 private wells in Montgomery County (2–20 identified PPCPs/well) 24 private wells in Roanoke County (2–17 identified PPCPs/well)	317
Oxcarbazepine	68% of wells				
Methylparaben	67% of wells				
Triclosan	61% of wells (0.78-53.55 ppb)				
Gabapentin	56% of wells				
Caffeine	51% of wells (0.69-1.53 ppb)				
Metformin	37% of wells				
Diphenhydramine	28% of wells				
Buprenorphine					
Tylosin	26% of wells (0.13-1.54 ppb)				
Paroxetine	23% of wells				
142 PPCPs were screened	16 were identified			6 municipal drinking water samples in Montgomery	

	in at least one samples			County (3), Roanoke County (2), and Botetourt County (1)	
Oxcarbazepine	in 5 samples				
<i>Triclosan</i>	in 5 samples (max = 22.9)				
Metformin	in 6 samples				
Caffeine	in 6 samples				
<i>Tylosin</i>	in 1 sample (max = 5.91)				

#Single value and value range in the table represent one sampling site and multiple sampling sites of a water body, respectively.

*References in bold italic font indicates studies cited in *Toxic Contaminants in the Chesapeake Bay and its Watershed: Extent and Severity of Occurrence and Potential Biological Effects – 2012 Technical Report*.¹

&Antibiotic compounds are in bold italic font.

ppb = parts per billion

Table A2. Occurrence of flame retardants in the waters of Virginia[#]						
Type of flame retardants	Water (ppb)	Sediment (ppb d.w.)	Organisms (ppb l.w.)		Locations	Ref*
PBDE congeners (BDE-47, 99, 100, 153, and 154)		42			Hyco River	318
		21			Roanoke River	
38 PBDE congeners		0.1	Mussels & oysters	78	Mattox Creek	319
				11	Chincoteague Inlet	
		82		Rappahannock River		
		106		Cape Charles		
		120		Sandy Point		
		0.4		170	James River	
PBDE congeners (BDE-47, 99, 100, 153, and 154)			Peregrine falcon eggs (ppb wet weight)	73	Chincoteague, Accomack County	320
				29	Fort Eustis, Newport News	
				91-131	James River, Newport News	
				111	Prince George County	
				109	Fisherman Island, Northampton County	
				178	Elkins Marsh, Northampton County	
				135-207	Berkeley Bridge, Norfolk	
				107	Norfolk Southern bridge, Norfolk	
29 PBDE congeners			Osprey eggs (ppb wet weight)	432	James River	321
				64	York River	
PBDE congeners (BDE-28, 47, 49, 99, 100, 153, 154, 183, and 209)			Common carp: 9140 Channel catfish: 1640 Redhorse sucker: 2270 Gizzard shad: 1120	Hyco River	94	
HBCD diastereomers (α -, β - and γ -HBCD)			Common carp: 4640 Channel catfish: 3680 Redhorse sucker: 1400 Gizzard shad: 290			
PBDE congeners (BDE-28, 47, 49, 99, 100, 153, 154, 183, and 209)			Common carp: 1180 Channel catfish: 600 Redhorse sucker: 600 Flathead catfish: 12450	Dan River		
HBCD diastereomers (α -, β - and γ -HBCD)			Common carp: 176 Channel catfish: 152 Redhorse sucker: 16 Flathead catfish: 698			
PBDE congeners (BDE-28, 47, 49, 99, 100, 153, 154, 183, and 209)			Common carp: 1340 Channel catfish: 160 Redhorse sucker: 580 Gizzard shad: 330	Roanoke River		
HBCD diastereomers (α -, β - and γ -HBCD)			Common carp: 54 Channel catfish: 60 Redhorse sucker: 21 Gizzard shad: 13			

[#]Single value and value range in the table represent average from one sampling site and multiple sampling dates/site or multiple sampling sites of a water body, respectively.

*References in bold italic font indicates studies cited in *Toxic Contaminants in the Chesapeake Bay and its Watershed: Extent and Severity of Occurrence and Potential Biological Effects – 2012 Technical Report*.¹

d.w. = dry weight; l.w. = lipid weight

Table A3. Occurrence of hormones in the waters of Virginia					
List of compounds identifiable or detectable (concentration range)					
Compounds	Water (ppb)	Sediment (ppb)	Organisms (intersex occurrence rate, %)	Locations	References*
17 α -ethynylestradiol	identifiable			Shenandoah River North Fork	<i>313</i>
17 β -estradiol equivalents (E2Eq) (estrogenic activity)	<0.0003 – 0.0072			Shenandoah River watershed (18 streams)	<i>115</i>
17 β -estradiol equivalents (E2Eq) (estrogenic activity)	0.004		Male smallmouth bass: 80-100	Shenandoah River North Fork	<i>322</i>
	0.005		Male smallmouth bass: 100	Shenandoah River South Fork	
17 α -estradiol 17 β -estradiol Androstenedione	detectable			Shenandoah River North Fork, Shenandoah River South Fork	<i>118</i>
17 β -estradiol	detectable	Max = 0.22			
Estrone	detectable	Max = 0.92			
Progesterone		Max = 6.38			
17 β -estradiol equivalents (E2Eq) (estrogenic activity)	detectable (quantitation limit = 0.0003 ppb)		Male largemouth bass: 22-38	Rappahannock River	<i>119</i>
			Male largemouth bass: 48	Motts Reservoir	
			Male largemouth bass: 0-30	Rappahannock River Valley (ponds)	
			Male largemouth bass: 60	Potomac River, Pohick Bay	
			Male largemouth bass: 100	Burke Lake	
			Male largemouth bass: 22	Back Bay (pond)	

*References in bold italic font indicates studies cited in *Toxic Contaminants in the Chesapeake Bay and its Watershed: Extent and Severity of Occurrence and Potential Biological Effects – 2012 Technical Report*.¹
ppb = parts per billion

Table A4. Occurrence of PFASs in the waters of Virginia							
Type of compound*	Water (ppt)	Sediment (ppb d.w.)	Organisms (ppb w.w.)	Locations		Ref	
PFOA+PFOS	41/43 wells: 1.26 – 493,600			Columbia Aquifer groundwater	Naval Air Station Oceana, Virginia Beach, Virginia	161	
PFBS	28/43 wells: detectable – 4,950						
PFHpA	31/47 wells: detectable – 13,900						
PFHxS	37/47 wells: detectable – 52,400						
PFNA	21/47 wells: detectable – 2,660						
PFOA+PFOS	5/6 well: 1.01-639.3			Yorktown Aquifer groundwater	161		
PFBS	1/6 well: detectable						
PFHpA	2/6 well: detectable -22.4						
PFHxS	3/6 well: detectable -124						
PFNA	1/6 well: detectable						
PFOA	Drinking water: 1,800			Naval Auxiliary Landing Field Fentress, Chesapeake, Virginia		184	
PFOS	Drinking water: 1,000						
PFOS			110-227	Elizabeth River (osprey eggs)			184
PFOA			NQ				
PFDS			NQ-41				
PFDA			NQ	Middle Potomac River (osprey eggs)			
PFOS			255-317				
PFOA			NQ-7.2				
PFDS			91-119				
PFDA			NQ-20.3				

*See Figure 5 for compound classification and acronym.

d.w. = dry weight basis; w.w. = wet weight basis; NQ = not quantifiable