Pharmaceuticals, Micropollutants and Antibiotics in Biosolids

38th Annual Spring Biosolids Symposium

Tuesday, March 9, 2021

MARQUETTE UNIVERSITY
BE THE DIFFERENCE.

Dr. Patrick McNamara

Marquette University

WATER QUALITY CENTER
MARQUETTE UNIVERSITY

Our Many Names For Pollutants

Contaminants of Emerging Concern

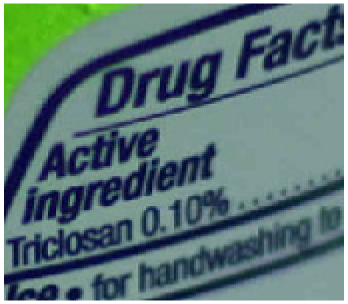
Micropollutants (organic micropollutants OMPs)

Trace Organic Contaminants (TOrCs)

Microconstituents

Pharmaceuticals and Personal Care Products (PPCP)

Where do they come from?



In the News – What Does it Mean?

AP: Drugs found in drinking water

Updated 9/12/2008 2:02 PM | Comments 153 | Recommend 97

By Jeff Donn, Martha Mendoza and

Drugs in Our Drinking Water?

Experts put potential risks in perspective after a report that drugs are in the water supply.

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Reviewed by Louise Chang,

-- including antibiotics, hormones, mood

46 million in U.S. have drugs in drinking water

Testing shows traces of meds in water gr

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Study finds hospitals hype robot surge

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Associated Press

updated 9/11/2008 4:29:08 PM ET

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Testing prompted by an
Press story that revealed
amounts of pharmaceuti
drinking water supplies

Tons of Released Drugs Taint U.S. Water

April 19, 2009 | RSS Feed | Print

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Painkiller Abuse by Kids Way Up, Study Finds

Tainted Steroid Injections May Affect Those Treated for Joint Pain

Associated Press

JEFF DONN

Associated Press Writers

U.S. manufacturers, including major drugmakers, have legally released at least 271 million pounds of pharmaceuticals into waterways that often provide drinking water — contamination the federal government has consistently overlooked, according to an Associated Press investigation.

Hundreds of active pharmaceutical ingredients are used in a variety of manufacturing, including drugmaking: For example, lithium is used to make ceramics and treat bipolar disorder; nitroglycerin is a heart drug and also used in explosives; copper shows up in everything from pipes to contraceptives.

Federal and industry officials say they don't know the extent to which pharmaceuticals are released by U.S. manufacturers

Seminal Study – They Are Everywhere

Environ. Sci. Technol. **2002**, *36*, 1202–1211

Pharmaceuticals, Hormones, and Other Organic Wastewater Contaminants in U.S. Streams, 1999–2000: A National Reconnaissance

DANA W. KOLPIN*
U.S. Geological Survey, 400 S. Clinton Street, Box 1230, Iowa City, Iowa 52244

EDWARD T. FURLONG
U.S. Geological Survey, Box 25046, MS 407, Denver, Colorado 80225-0046

MICHAEL T. MEYER
U.S. Geological Survey, 4500 SW 40th Avenue,

rarely exceeded drinking-water guidelines, drinking-water health advisories, or aquatic-life criteria. Many compounds, however, do not have such guidelines established. The detection of multiple OWCs was common for this study, with a median of seven and as many as 38 OWCs being found in a given water sample. Little is known about the potential interactive effects (such as synergistic or antagonistic toxicity) that may occur from complex mixtures of OWCs in the environment. In addition, results of this study demonstrate the importance of obtaining data on metabolites to fully understand not only the fate and transport of OWCs in the hydrologic system but also their ultimate overall effect on human health and the environment.

Introduction

The continued exponential growth in human population has created a corresponding increase in the demand for the

Seminal Study – They Are Everywhere

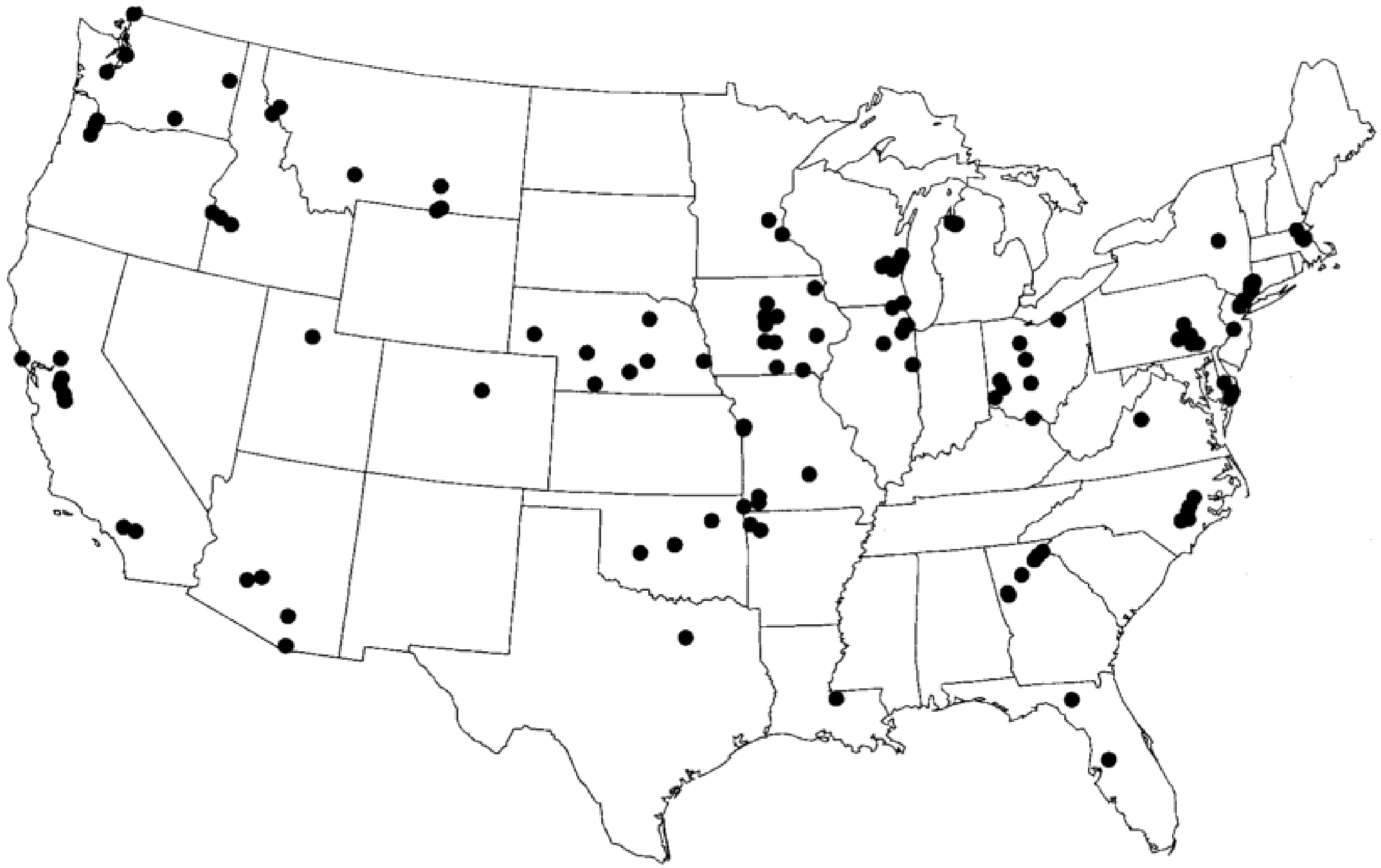


FIGURE 1. Location of 139 stream sampling sites.

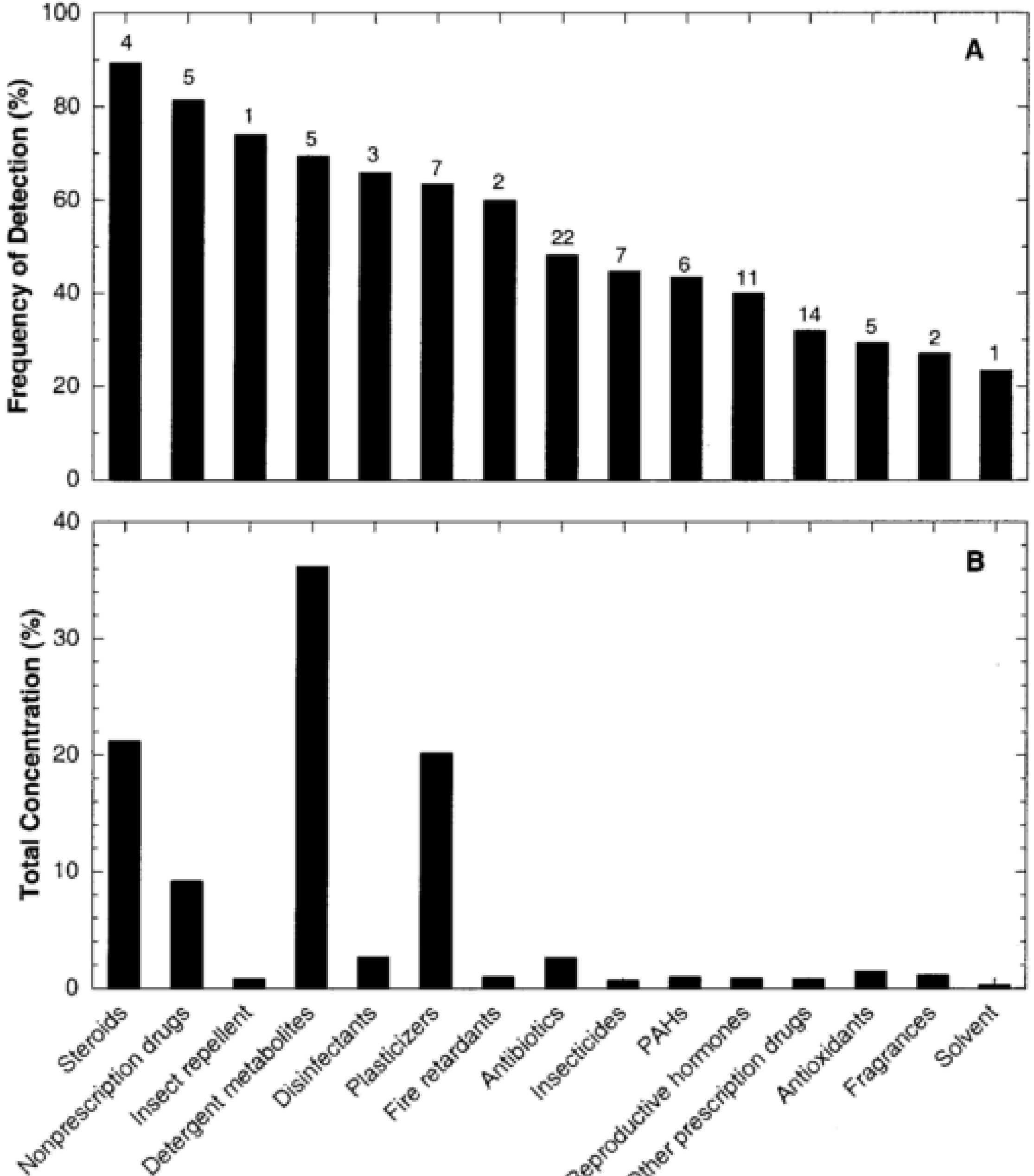
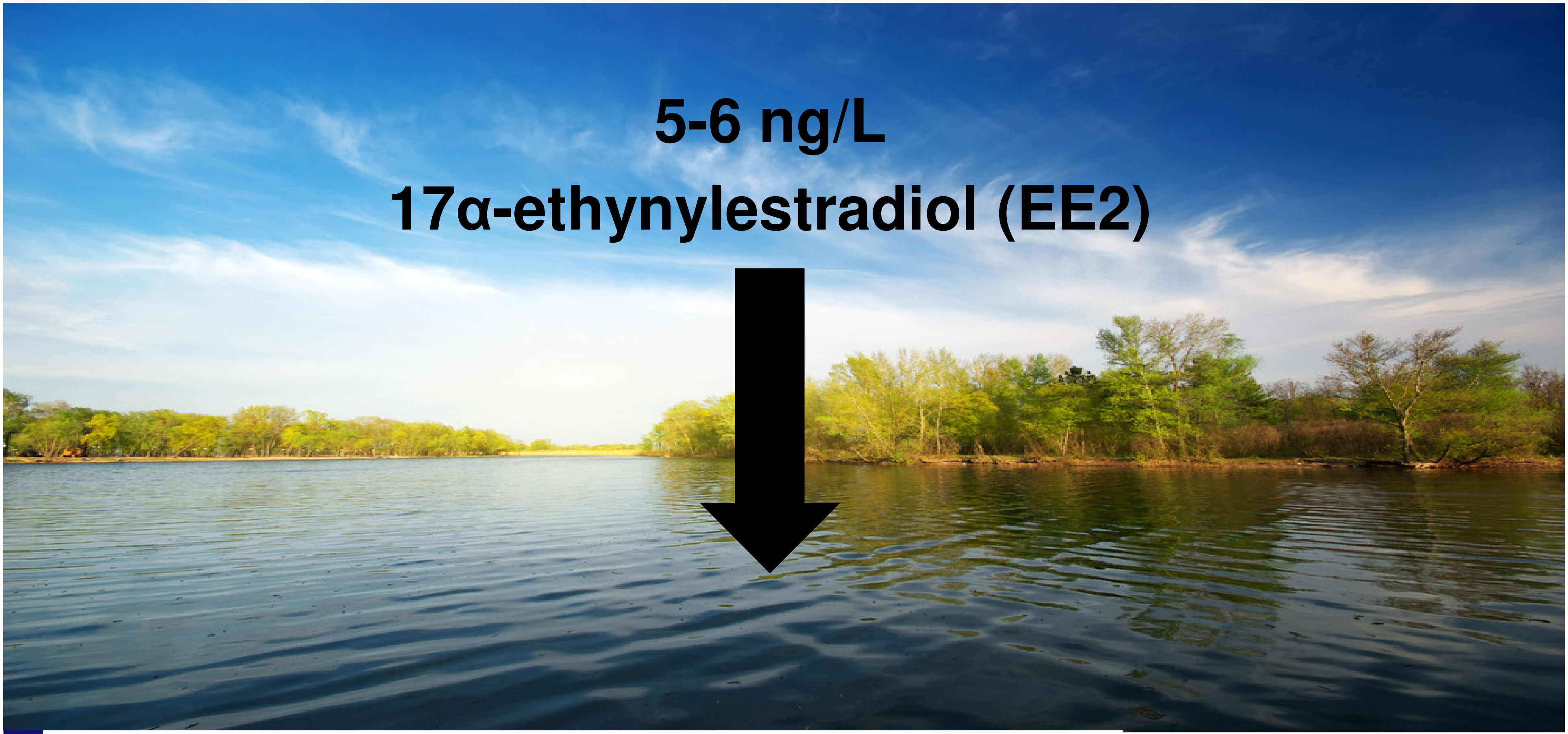


FIGURE 4. Frequency of detection of organic wastewater contaminants by general use category (4A), and percent of total measured concentration of organic wastewater contaminants by general use category (4B). Number of compounds in each category shown above bar.

Do they have any impact?

Lake Experiment – Spike in Realistic Dose



Collapse of a fish population after exposure to a synthetic estrogen

Karen A. Kidd*†, Paul J. Blanchfield*, Kenneth H. Mills*, Vince P. Palace*, Robert E. Evans*, James M. Lazorchak‡, and Robert W. Flick‡

*Fisheries and Oceans Canada, Freshwater Institute, 501 University Crescent, Winnipeg, Manitoba, Canada R3T 2N6; and ‡Molecular Indicators Research Branch, United States Environmental Protection Agency, 26 West Martin Luther King Drive, Cincinnati, OH 45268

Edited by Deborah Swackhamer, University of Minnesota, Minneapolis, MN, and accepted by the Editorial Board March 29, 2007 (received for review October 27, 2006)

PNAS

Evidence for Fish Feminization

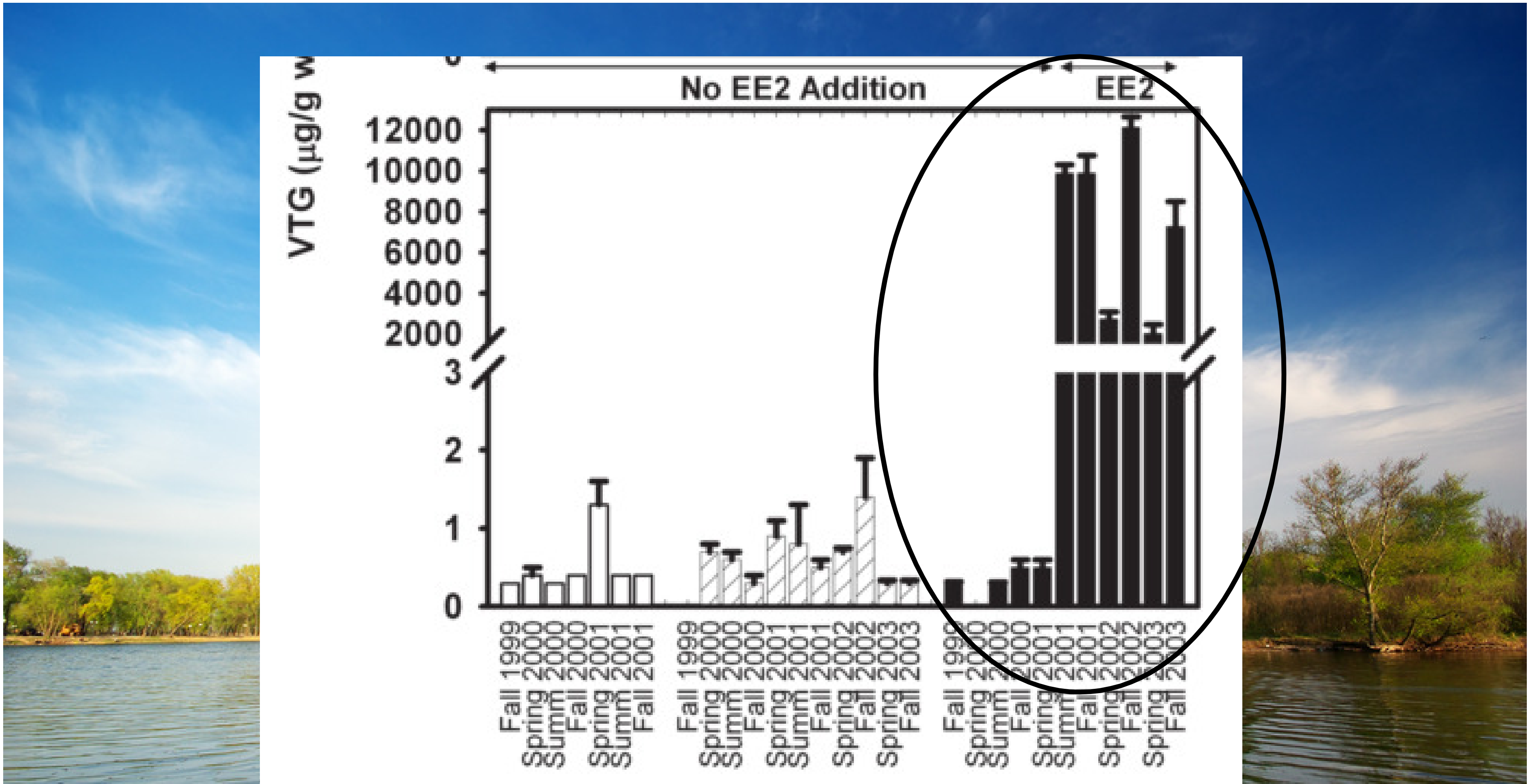


Fig. 1. Mean ± SE (n = 4–7) VTG concentrations in whole-body homogenates of male (*Lower*) and female (*Upper*) fathead minnow captured in 1999–2003 from reference Lakes 114 and 442 and from Lake 260 before and during additions of 5–6 ng·L^{-1} of EE2 (low catches of fish in Lake 260 in 2004 and 2005 did not allow for these analyses in the latter 2 years of the study).

Eventual Collapse of Population

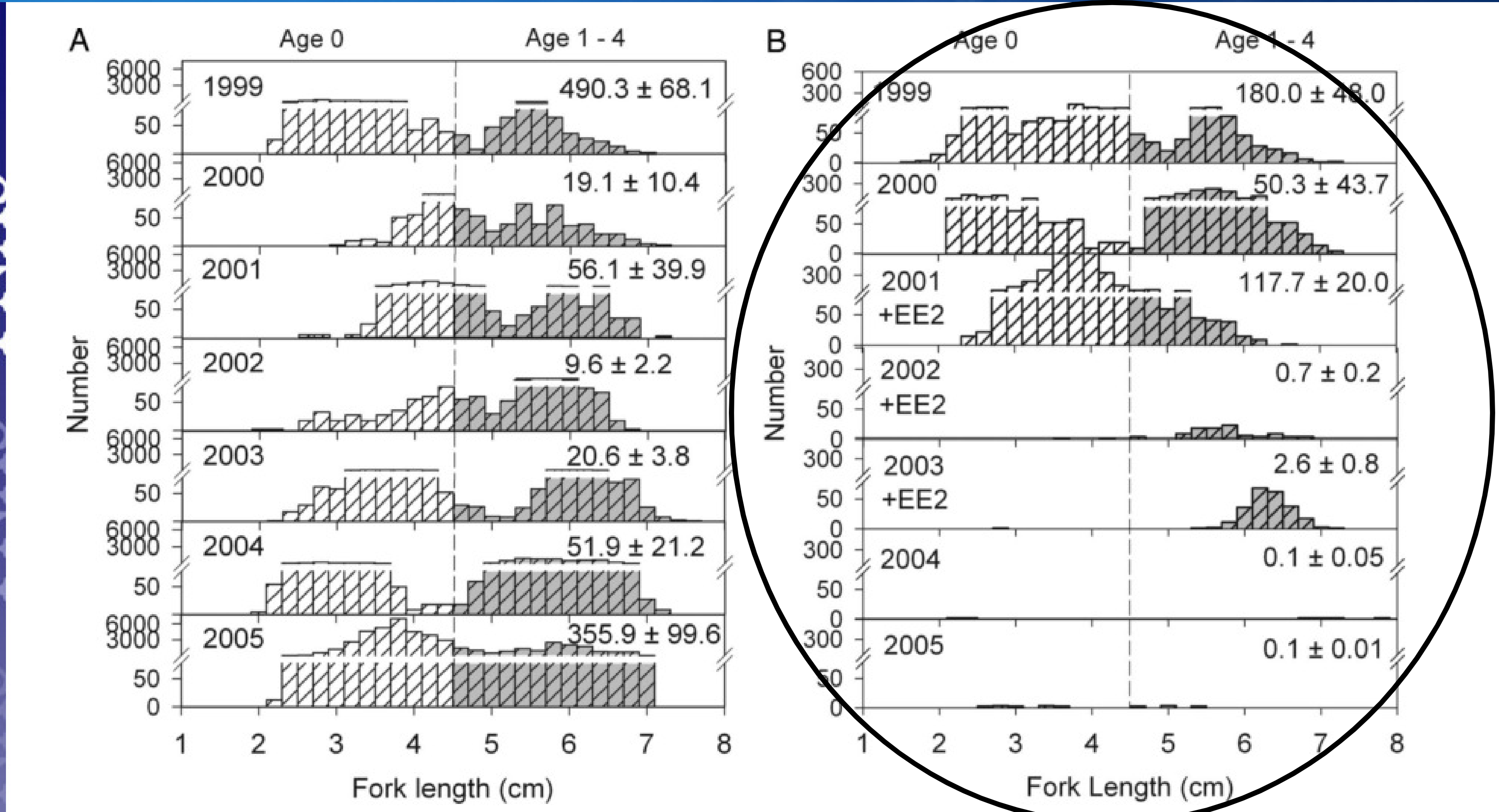
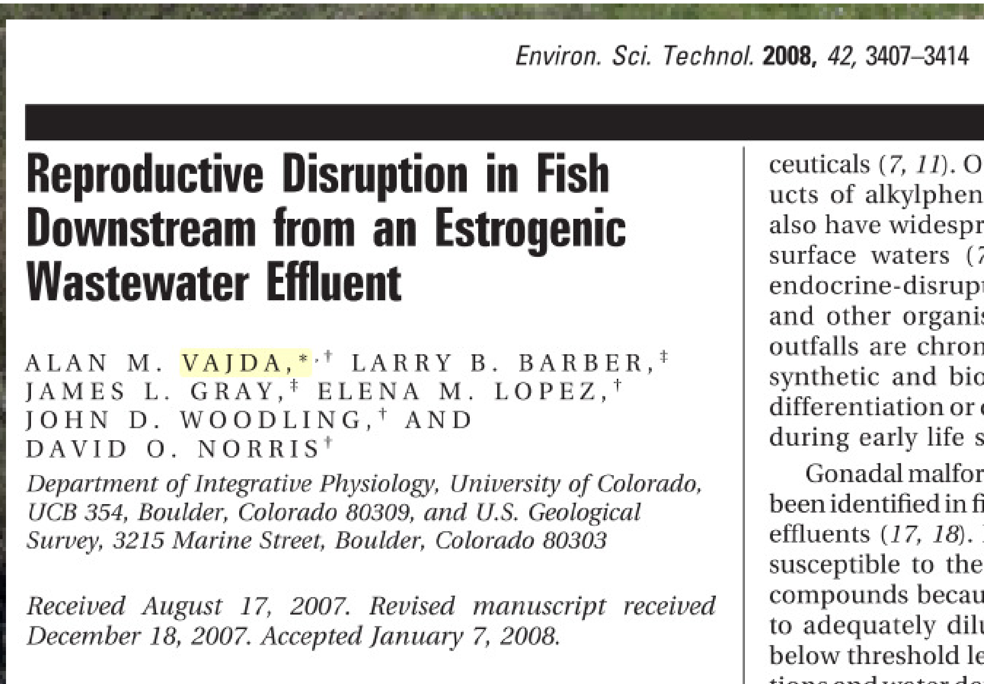


Fig. 3. Length frequency distributions of fathead minnow captured in trap nets in reference Lake 442 (*A*) and Lake 260 (*B*) (amended with 5–6 ng·L^{-1} of EE2 in 2001–2003) during the fall of 1999–2005. Distributions for each fall have been standardized to 100 trap-net days. Mean ± SE daily trap-net CPUE data for adults and juveniles for the fall catches are shown in the panels.

Nice experiment, but are there any real world impacts?

Wastewater Influences Minnow Gender



Environ. Sci. Technol. **2008,** *42,* 3407–3414

Reproductive Disruption in Fish Downstream from an Estrogenic Wastewater Effluent

ALAN M. VAJDA,*,† LARRY B. BARBER,‡ JAMES L. GRAY,‡ ELENA M. LOPEZ,† JOHN D. WOODLING,† AND DAVID O. NORRIS†

Department of Integrative Physiology, University of Colorado, UCB 354, Boulder, Colorado 80309, and U.S. Geological Survey, 3215 Marine Street, Boulder, Colorado 80303

Received August 17, 2007. Revised manuscript received December 18, 2007. Accepted January 7, 2008.

ceuticals (*7, 11*). Ot
ucts of alkylphen
also have widespr
surface waters (*7*
endocrine-disrupt
and other organis
outfalls are chron
synthetic and bio
differentiation or c
during early life s

Gonadal malfor
been identified in fi
effluents (*17, 18*). I
susceptible to the
compounds becau
to adequately dilu
below threshold le

Upstream of WWTP

36-46% Males

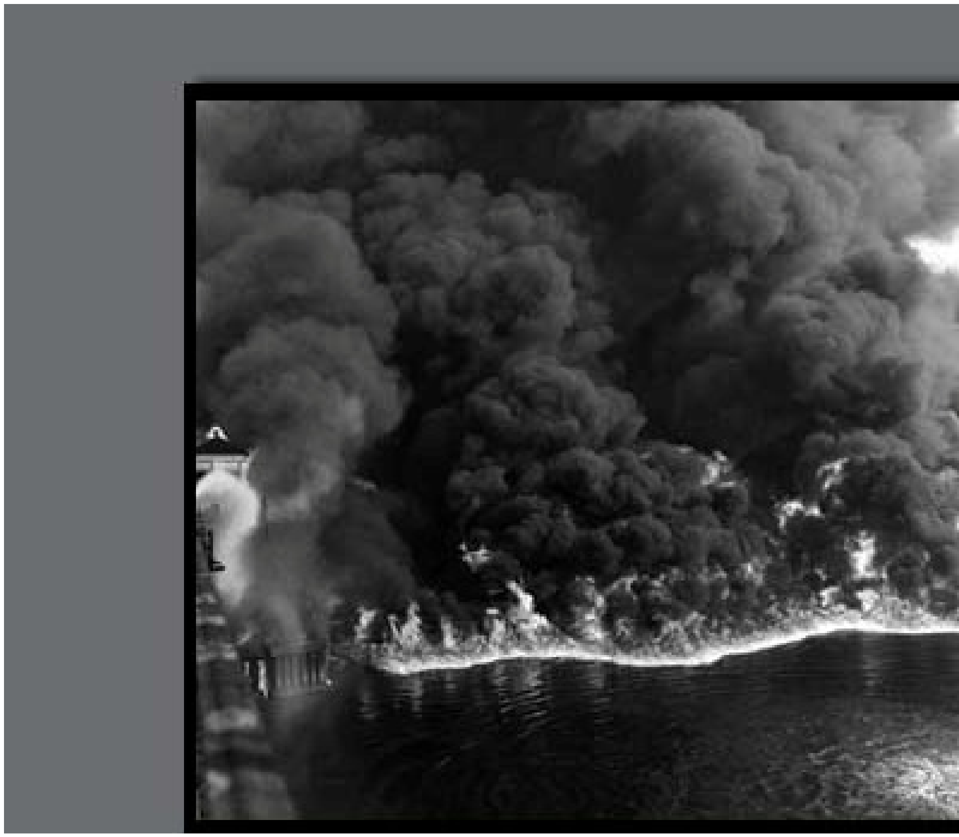
Downstream of WWTP

17-21% Males
18-22% Intersex

Micropollutants, with Designed Biological Impacts,

Impact Biological Systems

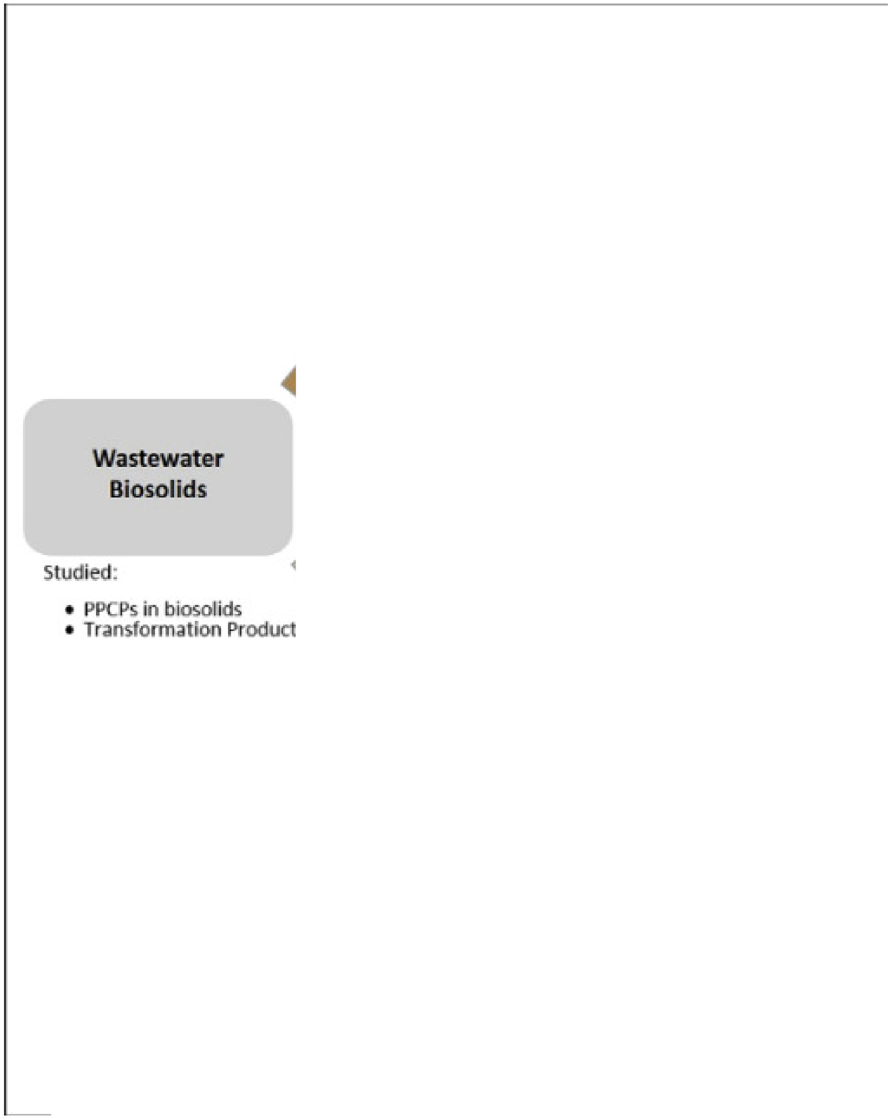
Acute Toxicity → Chronic Low-level exposure



What about micropollutants in biosolids?

Translocation of pharmaceuticals and personal care products after land application of biosolids

Chad A. Kinney[1] and Brian Vanden Heuvel[2]



Highlights of current understanding related to PPCPs in biosolids and plant uptake of PPCPs along with key knowledge gaps identified.

What micropollutants are in biosolids?

WATER RESEARCH 44 (2010) 658–668

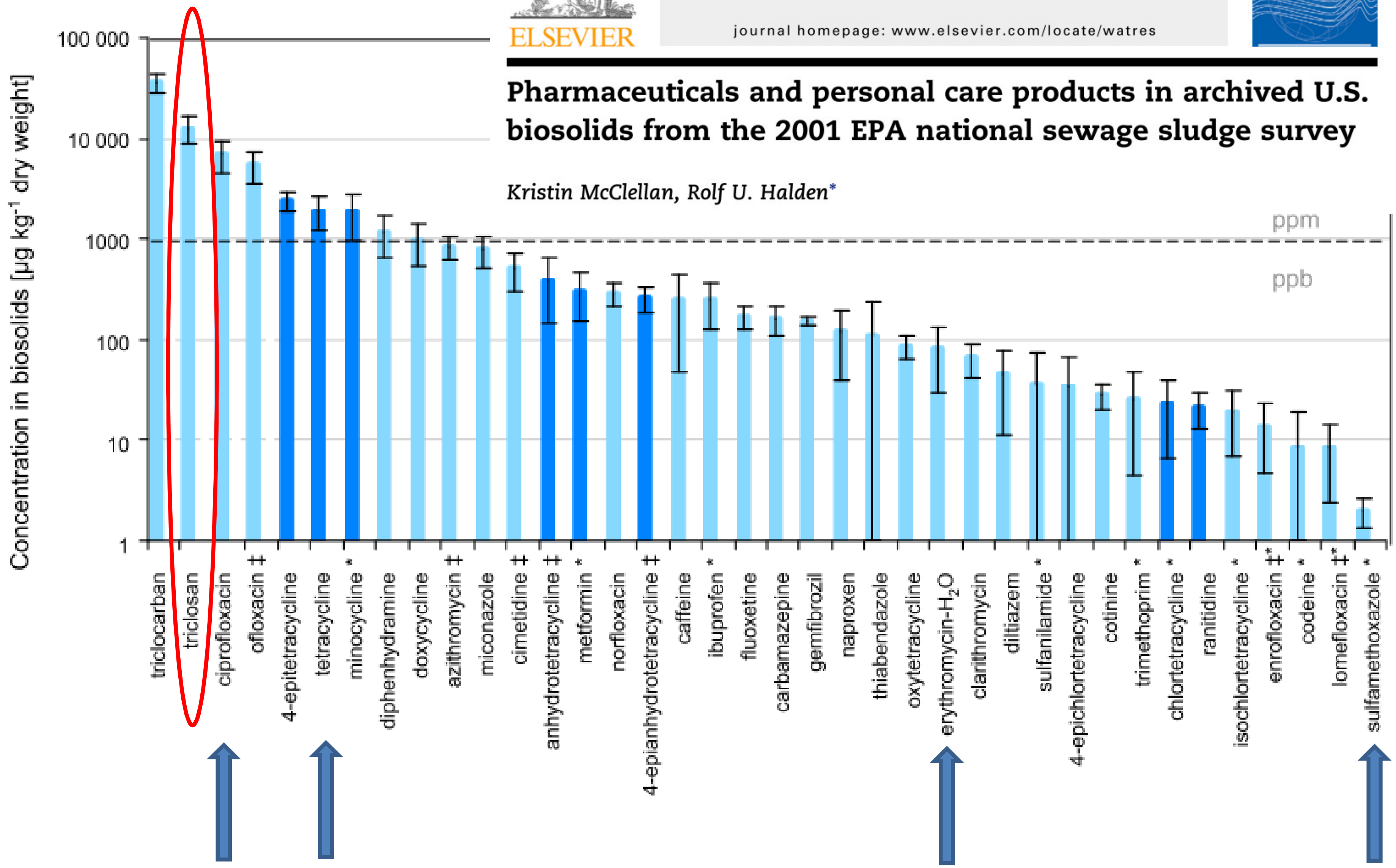
Available at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/watres

Pharmaceuticals and personal care products in archived U.S. biosolids from the 2001 EPA national sewage sludge survey

*Kristin McClellan, Rolf U. Halden**



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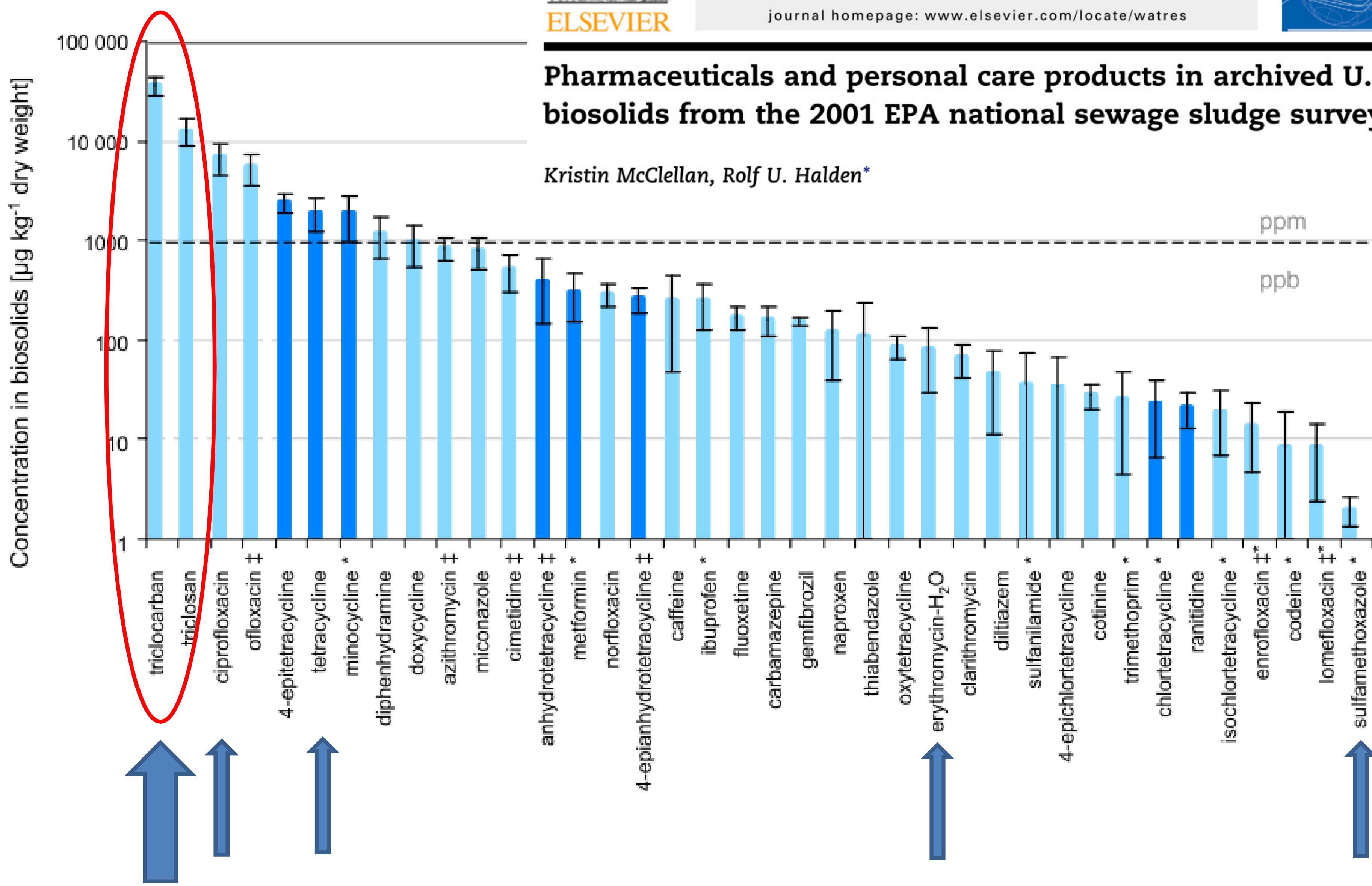


The Broad Research Question

Do triclosan and triclocarban select for antibiotic resistance genes in anaerobic digestion?

Why Do We Care?

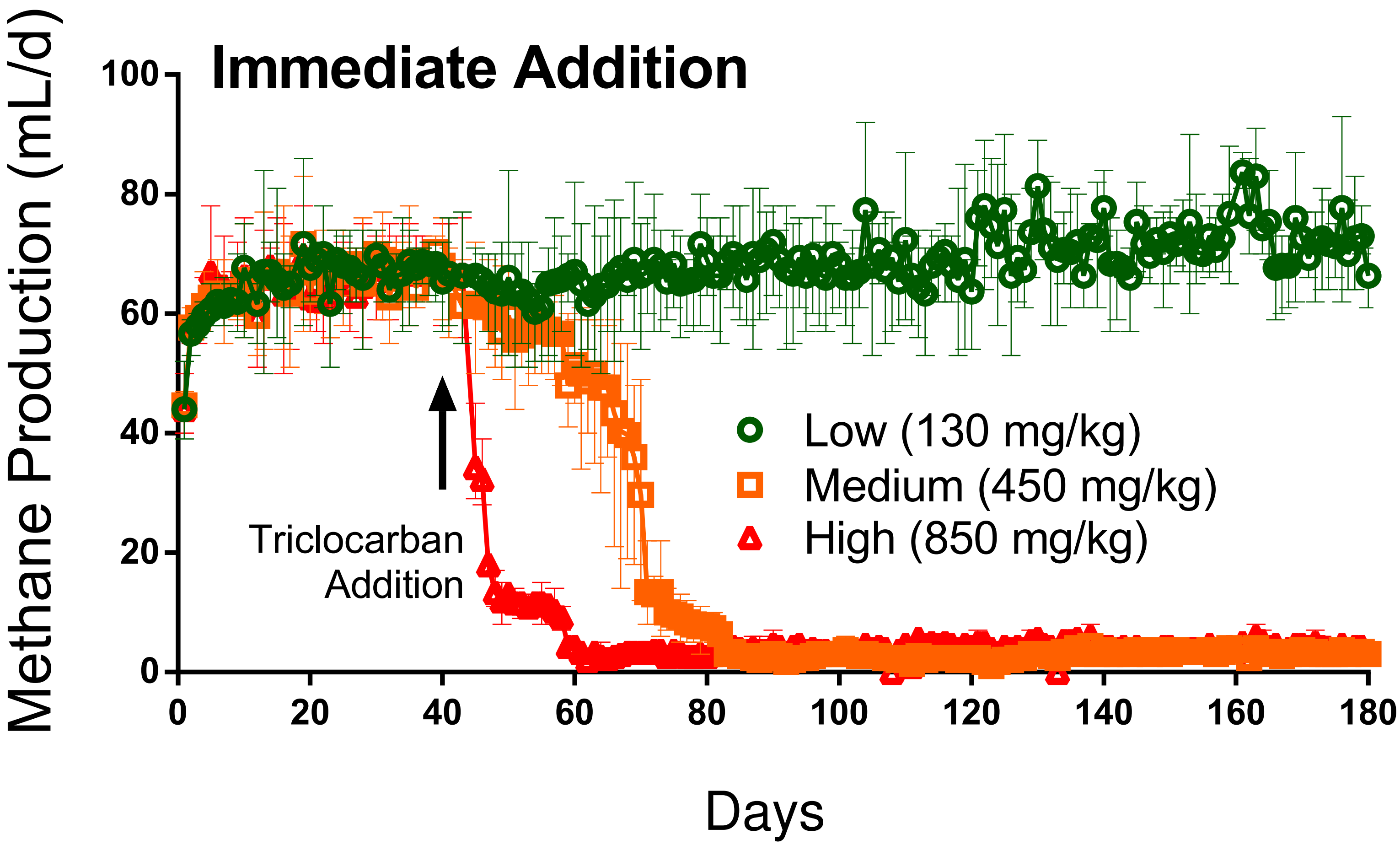
Antibiotic resistance genes could subsequently be released to the environment with biosolids

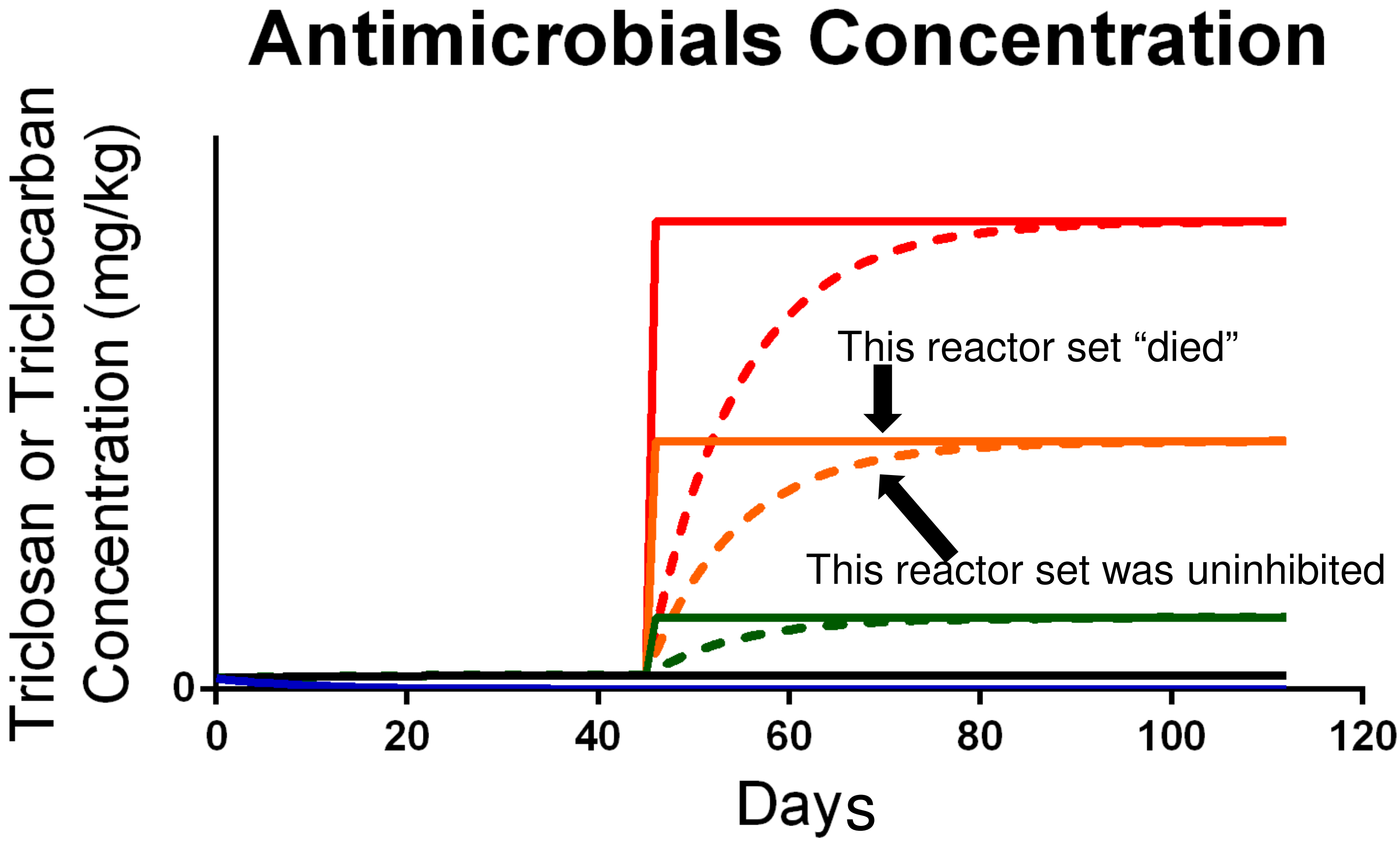
Research Approach

Lab Scale Digesters Amended with TCC

Carey et al., *Environ. Sci. Technol.* 2016, 50 (1), 126-134

Methane Production - Triclocarban

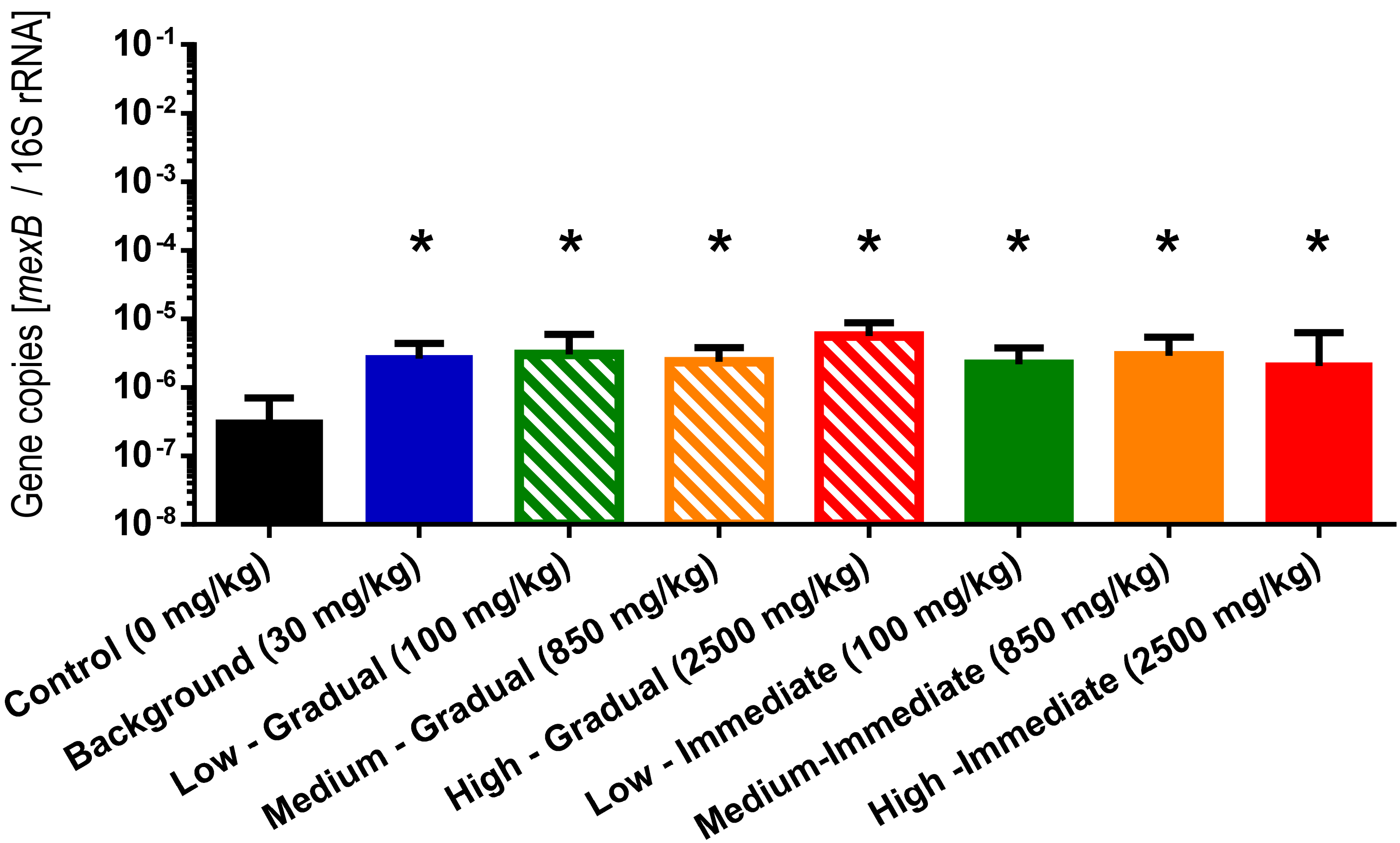




Antimicrobials Concentration
Triclosan or Triclocarban Concentration (mg/kg)
0
0
20
40
60
80
100
120
Days
This reactor set "died"
This reactor set was uninhibited

Antibiotic Resistance Genes

mexB increases from TCS



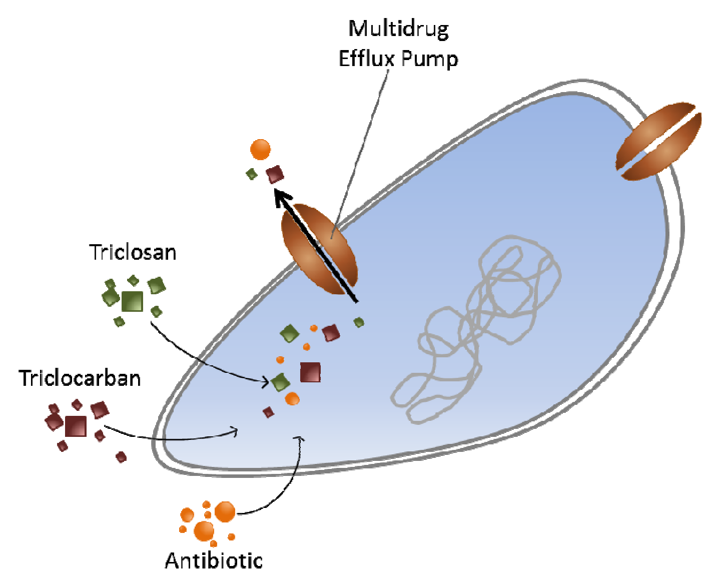
Carey et al., *Env. Sci: Proc. & Imp*. 2016, 18, 1060-67

Key Finding:

Triclosan and triclocarban can select for antibiotic resistance genes at environmentally relevant levels in anaerobic digesters

Implications

- Consumer product chemicals are likely already selecting for resistance in treatment systems
  - TCS and TCC selected for a multidrug resistance gene at concentrations between 30-850 mg/kg



FDA bans Triclosan and Triclocarban from soaps

U.S. Department of Health and Human Services

FDA U.S. FOOD & DRUG ADMINISTRATION

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FDA News Release

FDA issues final rule on safety and effectiveness of antibacterial soaps

Rule removes triclosan and triclocarban from over-the-counter antibacterial hand and body washes

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For Immediate Release September 2, 2016

Release

Español

The U.S. Food and Drug Administration today issued a final rule establishing that over-the-counter (OTC) consumer antiseptic wash products containing certain active ingredients can no longer be marketed. Companies will no longer be able to market antibacterial washes with these ingredients because manufacturers did not demonstrate that the ingredients are both safe for long-term daily use and more effective than plain soap and water in preventing illness and the spread of certain infections. Some manufacturers have already started removing these ingredients from their products.

Inquiries

Media

Andrea Fischer

Consumers

888-INFO-FDA

Related Information

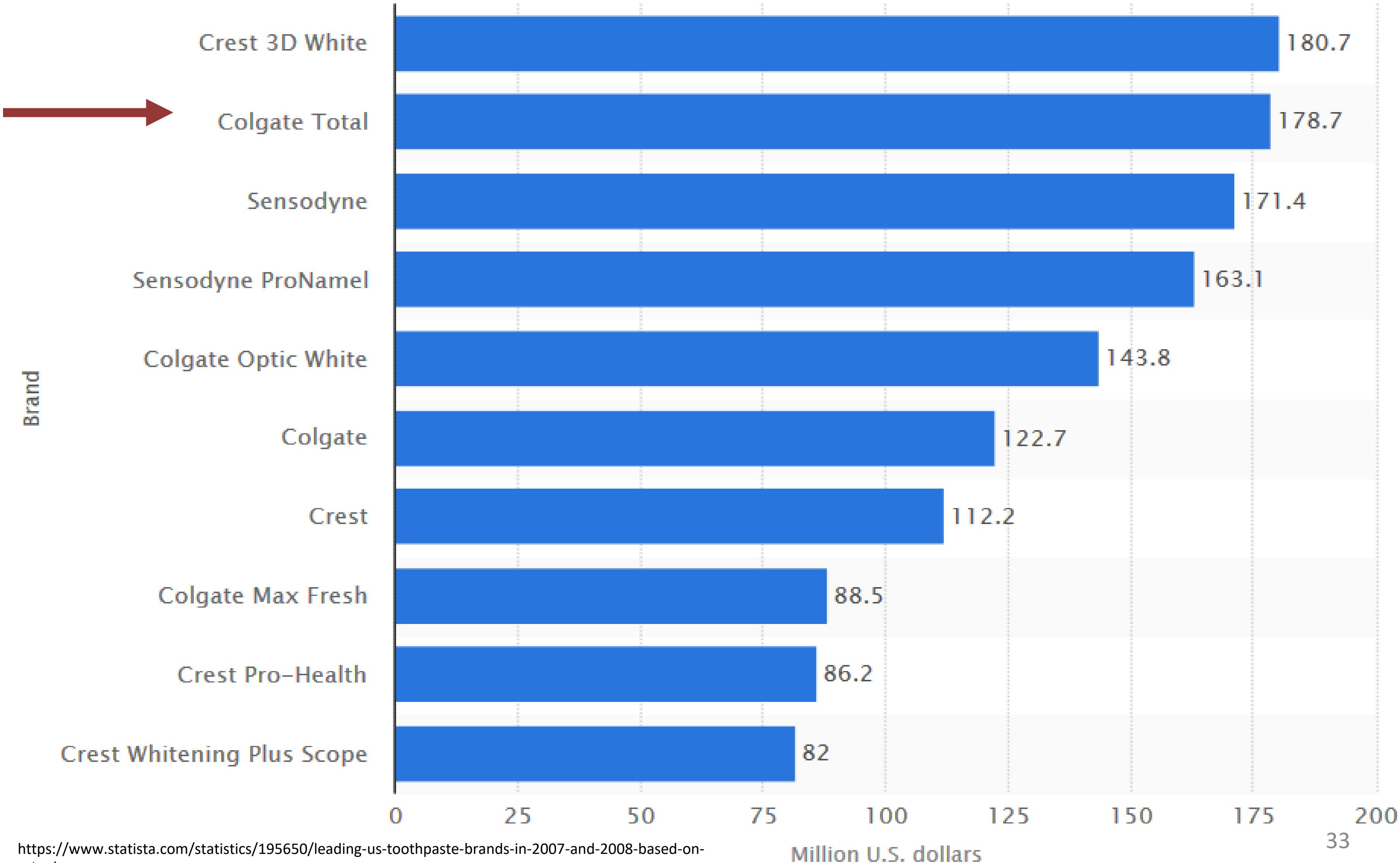
- Final Rule: Safety and Effectiveness of Consumer Antiseptics
- Antibacterial Soap? You Can Skip It -- Use Plain Soap and Water
- Topical Antiseptic Products
- Handwashing: Clean Hands Save Lives (CDC)

http:/ .htm

What next?


Anticavity Fluoride
ADA Accepted
NET WT 6.0 OZ (170 g)
HELPS
Drug Facts
Sodium Fluoride 0.24% (0.14% w/v fluoride io
Triclosan 0.30%
aids in the prevention of:
• cavities
of periodontitis, a ser
form of gum disease.

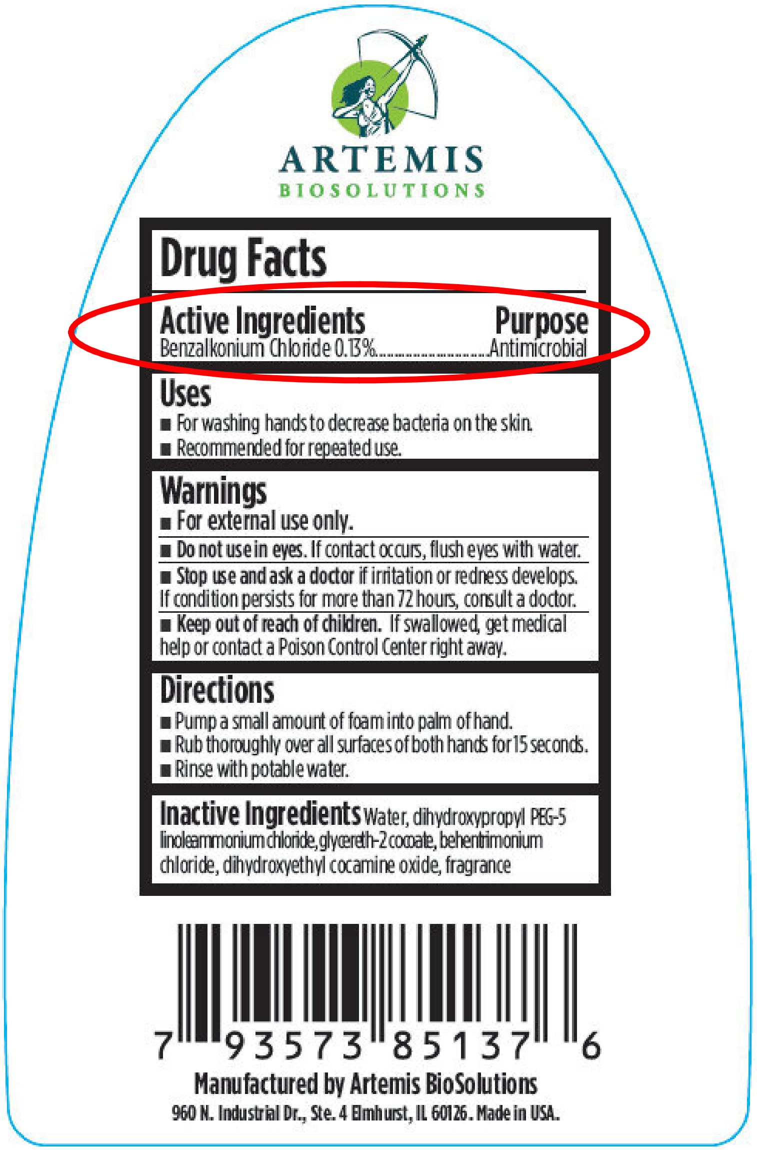
Triclosan Still in Top-Selling Toothpaste



https://www.statista.com/statistics/195650/leading-us-toothpaste-brands-in-2007-and-2008-based-on-sales/

Triclosan Free Soap

Benzalkonium Chloride



Science & Technology LETTERS
pubs.acs.org/journal/estlcu
Review

Increased Use of Quaternary Ammonium Compounds during the SARS-CoV-2 Pandemic and Beyond: Consideration of Environmental Implications

Priya I. Hora, Sarah G. Pati, Patrick J. McNamara, and William A. Arnold*

Benzalkonium Chloride: Limiting Stressors?

Environmental Science & Technology

Article

pubs.acs.org/est

Long-Term Exposure to Benzalkonium Chloride Disinfectants Results in Change of Microbial Community Structure and Increased Antimicrobial Resistance

Madan Tandukar,[†] Seungdae Oh,[†] Ulas Tezel,[†,§] Konstantinos T. Konstantinidis,[†,‡] and Spyros G. Pavlostathis*[,†]

[†]School of Civil and Environmental Engineering and [‡]School of Biology, Georgia Institute of Technology, Atlanta Georgia 30332-0512, United States

[§]The Institute of Environmental Sciences, Bogazici University, Bebek, Istanbul, 34342, Turkey

MU Research

Environmental Pollution 257 (2020) 113472

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

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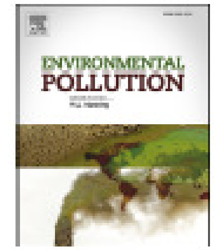
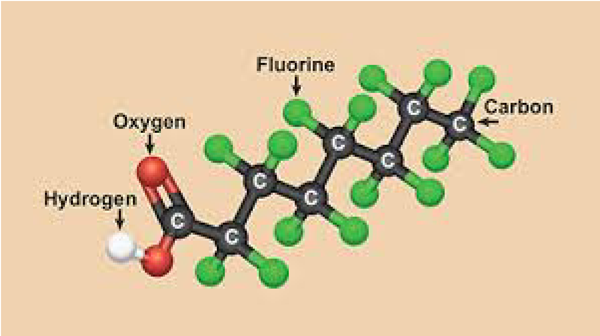


Benzalkonium chloride alters phenotypic and genotypic antibiotic resistance profiles in a source water used for drinking water treatment☆

Katherine R. Harrison [1], Anthony D. Kappell [2], Patrick J. McNamara*

Department of Civil, Construction and Environmental Engineering, Marquette University, Milwaukee, WI, USA

Updates on PFAS



Journal of Hazardous Materials 252–253 (2013) 413–418

Contents lists available at SciVerse ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

National inventory of perfluoroalkyl substances in archived U.S. biosolids from the 2001 EPA National Sewage Sludge Survey

Arjun K. Venkatesan[a], Rolf U. Halden[a,b,*]

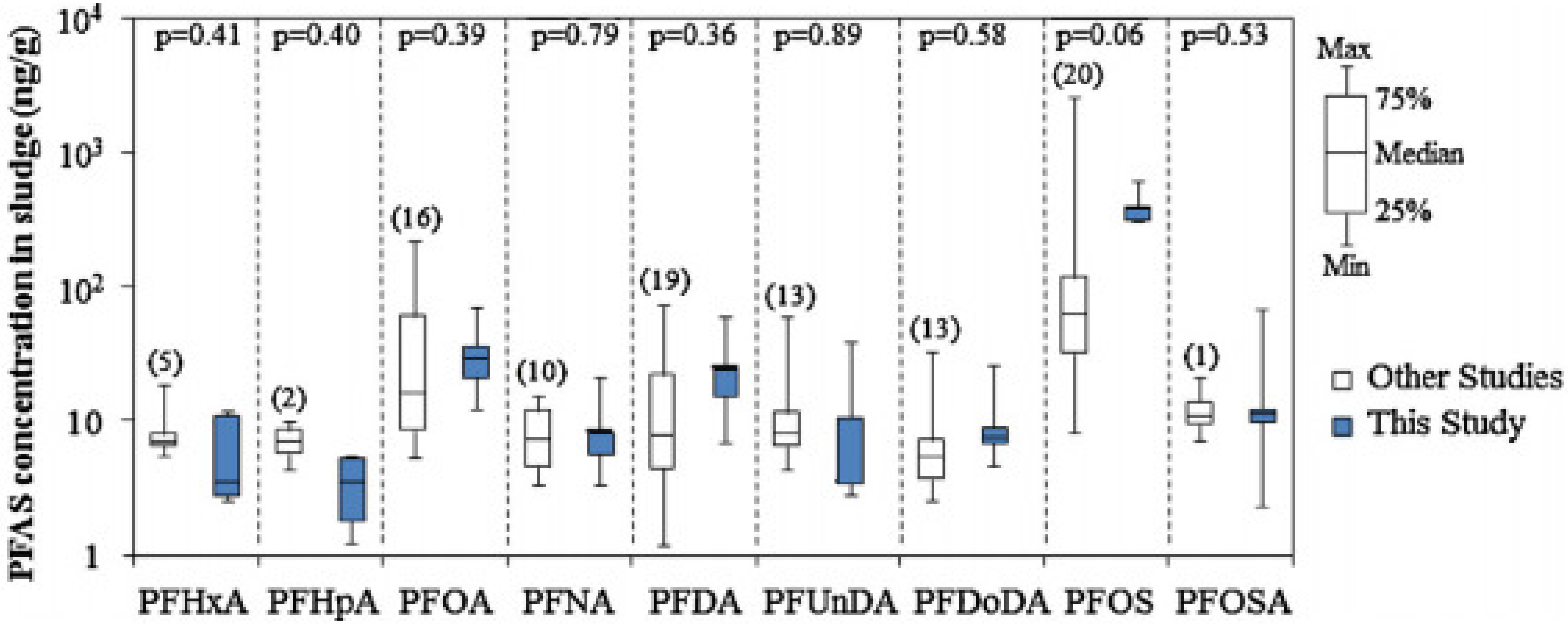
[a] Center for Environmental Security, The Biodesign Institute, Security and Defense Systems Initiative, Arizona State University, 781 E. Terrace Road, Tempe, AZ 85287, USA

[b] Department of Environmental Health Sciences, Bloomberg School of Public Health, Johns Hopkins University, Baltimore, MD, USA

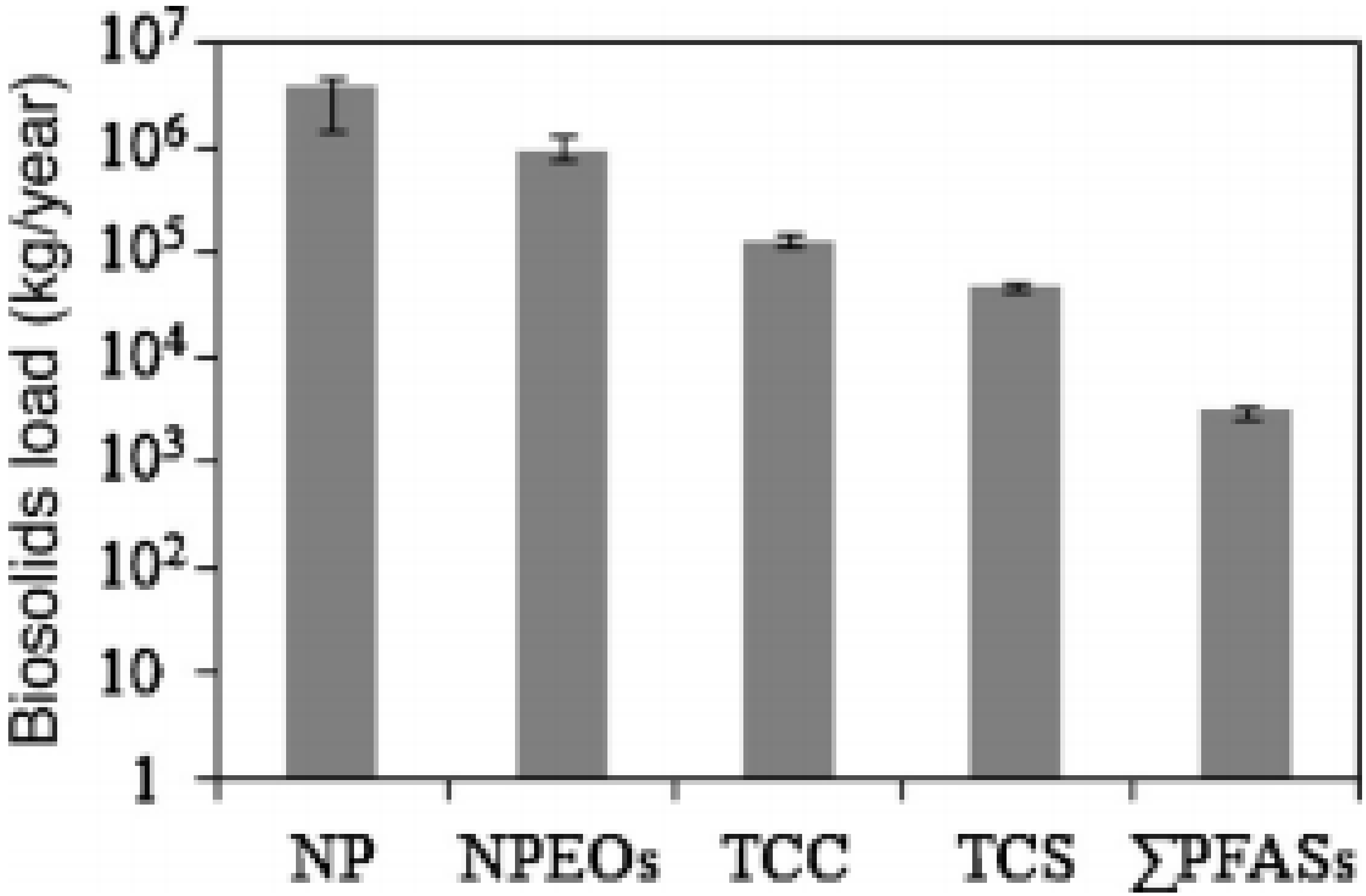
HIGHLIGHTS

- First study to report nationwide occurrence and concentrations of perfluoroalkyl substances (PFAS) in U.S. biosolids.
- Ten out of thirteen PFAS analyzed were consistently detected in all biosolids samples.
- PFOS was the most abundant PFAS in biosolids, followed by PFOA.
- Mean load of $\sum$PFASs in U.S. biosolids was estimated at 2749–3450 kg/year.
- PFASs in biosolids show no significant difference between pre- and post-phase out period.

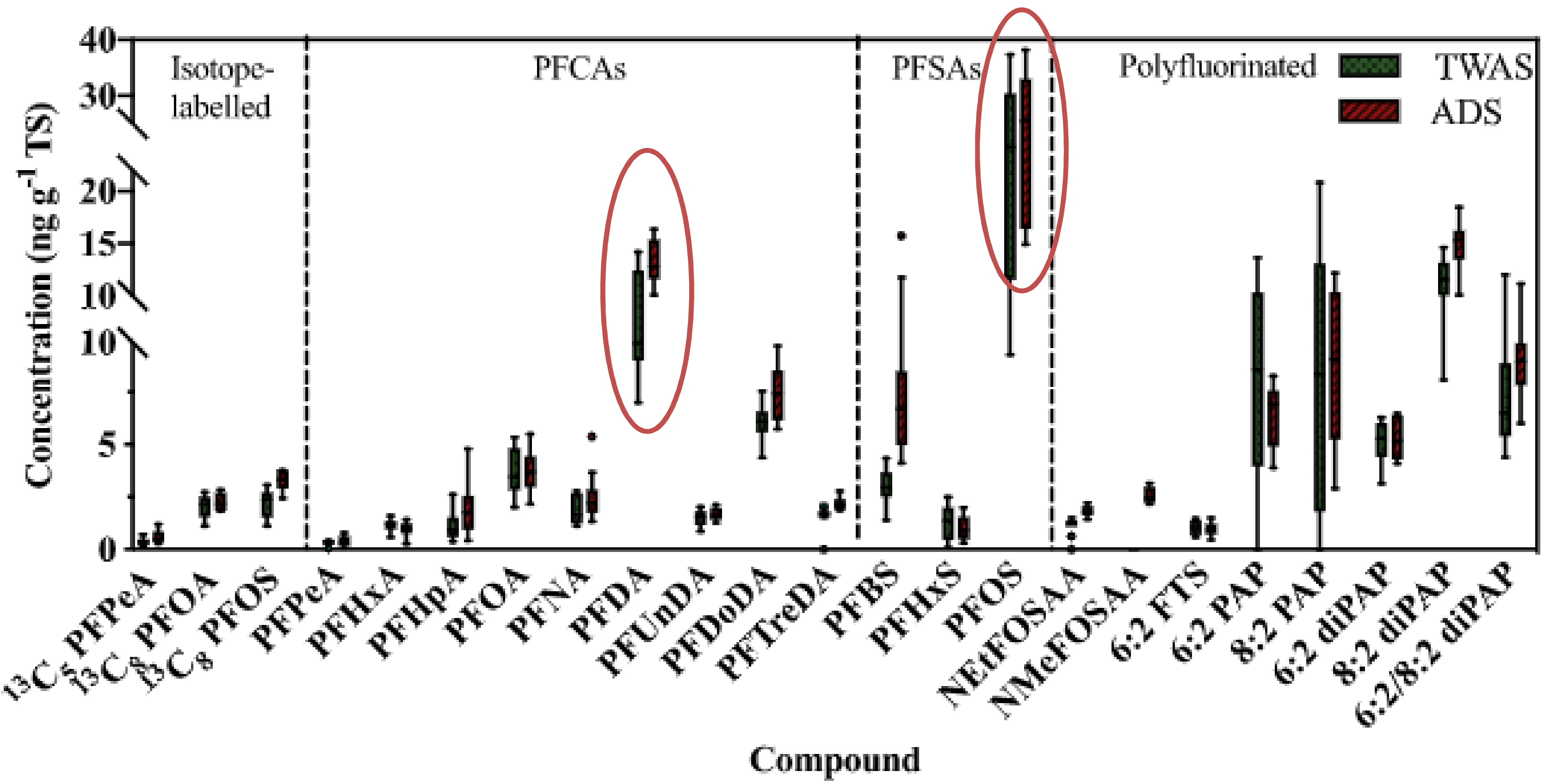
PFAS found at ng/g in biosolids



PFAS annual load less than other micropollutants



Short-chain PFAAs (C5 –C7 PFCAs and PFBS (C4)) can be generated from their precursors in AD



Formation and partitioning behaviour of perfluoroalkyl acids (PFAAs) in waste activated sludge during anaerobic digestion

Yijing Li[a], Jennifer Bräunig[b], Guerrero C. Angelica[a], Phong K. Thai[b], Jochen F. Mueller[b], Zhiguo Yuan[a,*]

Per- and polyfluoroalkyl substances in commercially available biosolid-based products: The effect of treatment processes

Rooney Kim Lazcano,[1,2,*] Chloé de Perre,[1] Michael L. Mashtare,[1,2,3] Linda S. Lee[1,2,3]

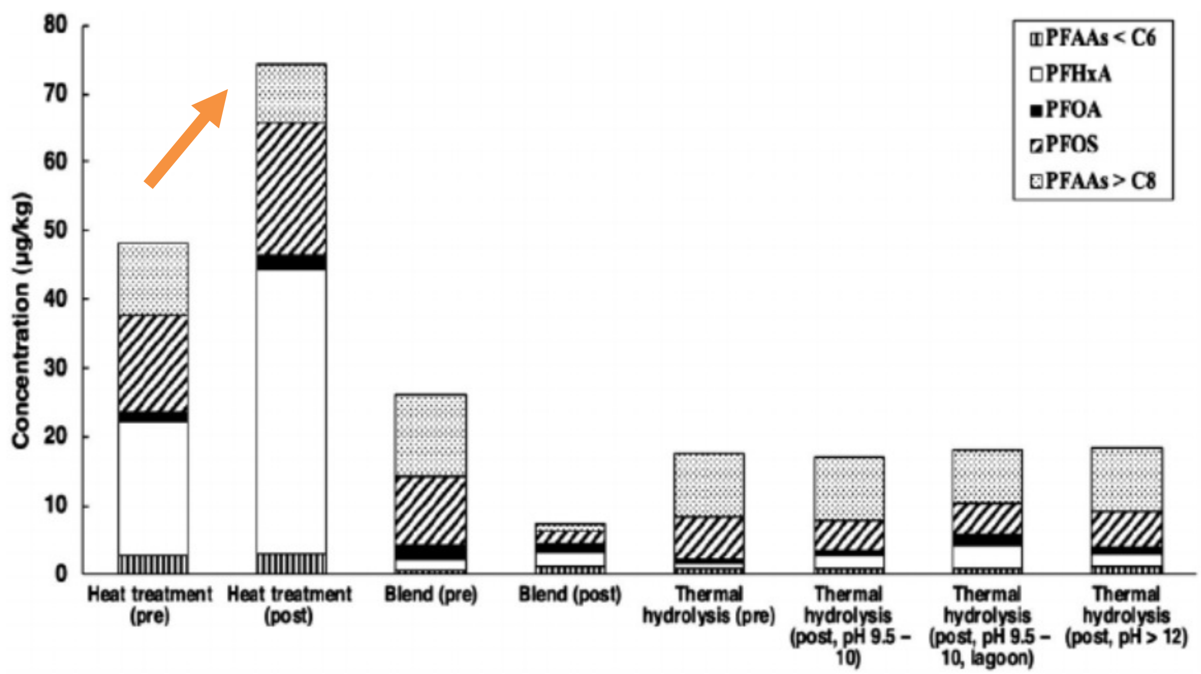


Figure 1. PFAA loads (µg/kg, dry wt.) for the <2 mm particle size fraction of the samples. Pre: before post-treatment process (the Class A or B biosolids) and post: after post-treatment process. PFAAs <C6 include PFBA and PFBS, and PFAAs >C8 include PFNA, PFDA, PFUdA, PFDoA, PFTrDA, and PFTeDA.

Influent PFAS Load Matters

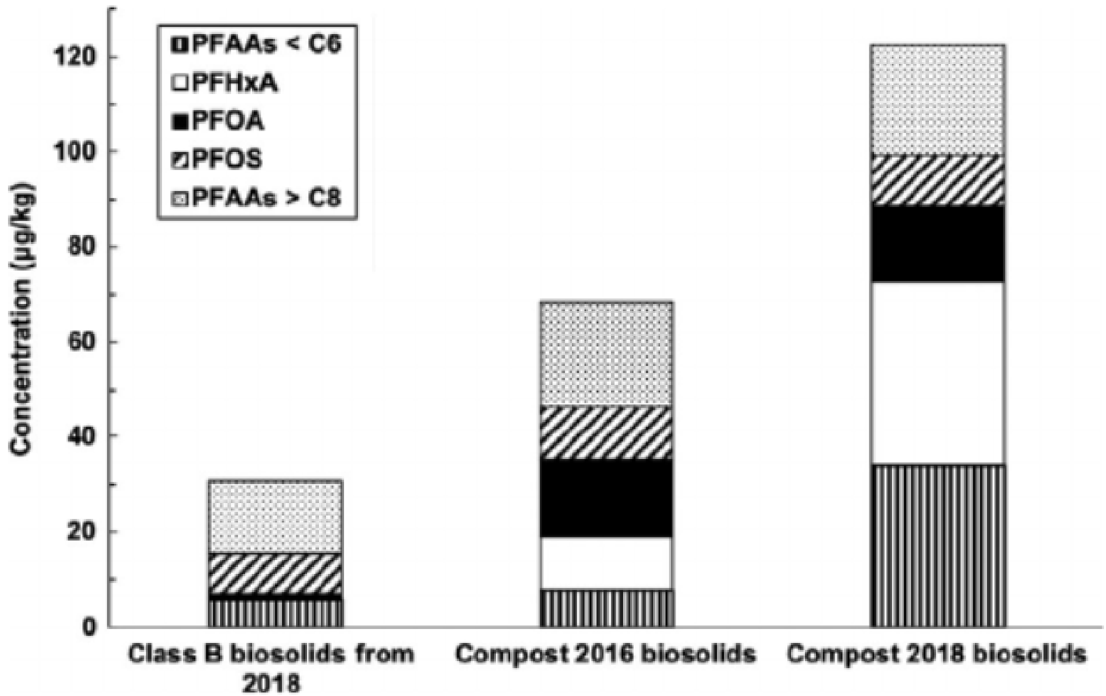


Figure 4. PFAA loads (µg/kg, dry wt.) for the <2 mm particle size fraction of the Class B biosolids from 2018 from one municipal water resource recovery facility (WRRF) and final composted 2016 and 2018 fertilizer products that contained Class B biosolids from four different WRRFs. Only one source of the Class B biosolids from 2018 was obtained and analyzed. PFAAs <C6 include PFBA and PFBS, and PFAAs > C8 include PFNA, PFDA, PFUdA, PFDoA, PFTrDA, and PFTeDA.

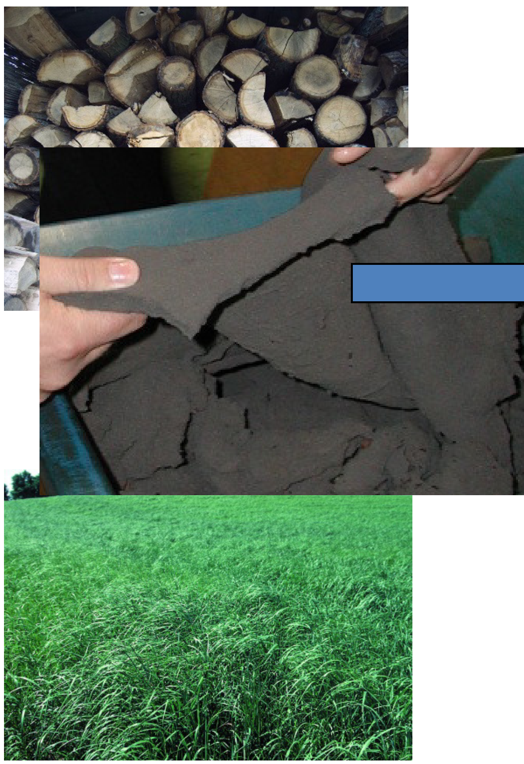
- **Practitioner points**
  - Heat treatment and composting increased perfluoroalkyl acid (PFAA) concentrations.
  - Only dilution from blending with non-PFAS material decreased PFAA concentrations.
  - Thermal hydrolysis process had no apparent effect on PFAA concentrations.
  - PFAS sources are a greater driver of PFAS loads in biosolid-based products than treatment processes.

We know micropollutants are in biosolids – are there technologies to get rid of them?

Pyrolysis: Heating Without Oxygen

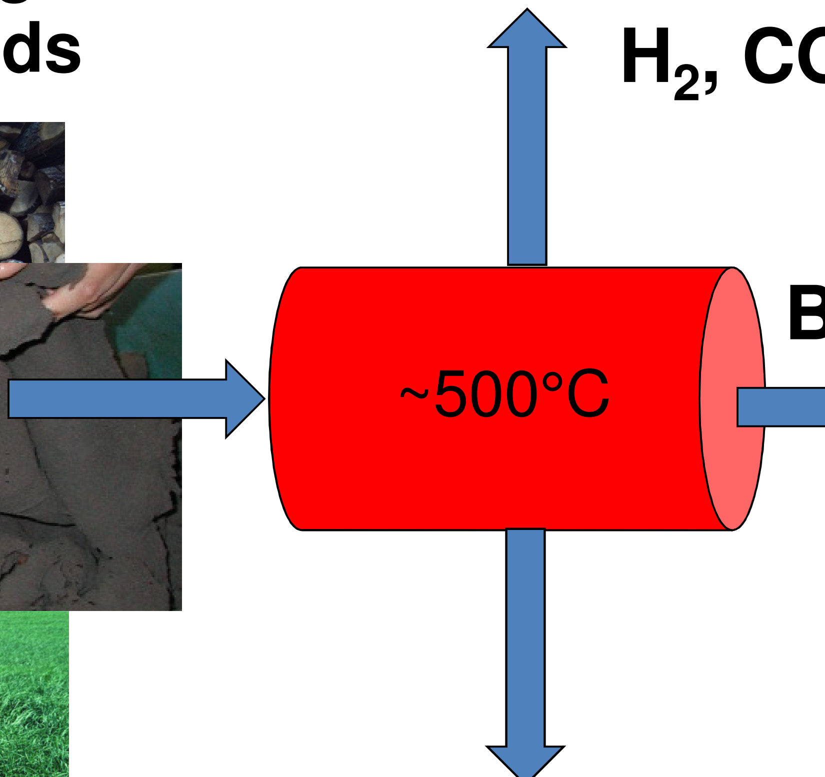
Biomass

- Wood
- Switchgrass
- Biosolids



Py-gas (5-30%)

H_2, CO, CH_4

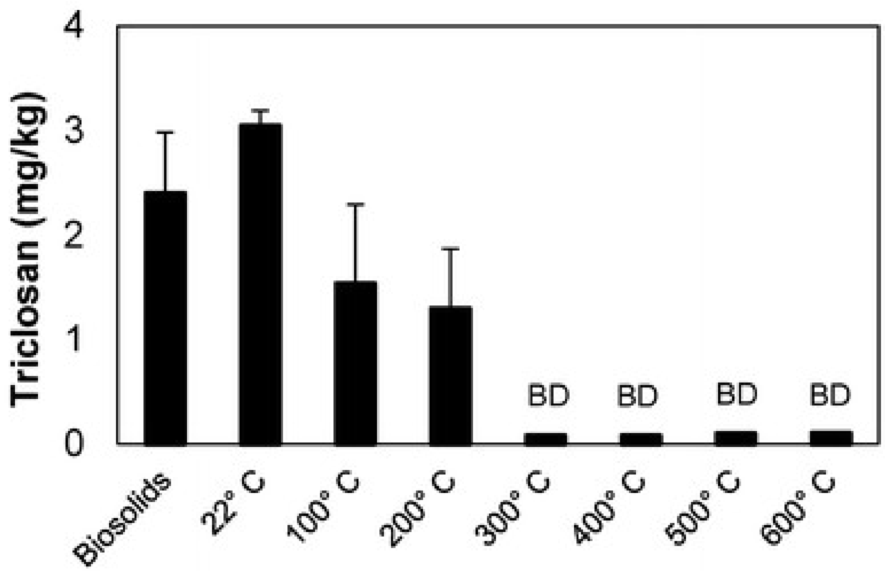


Bio-oil (20-50%)

Biochar (~50%)

Pyrolysis removes micropollutants from biosolids

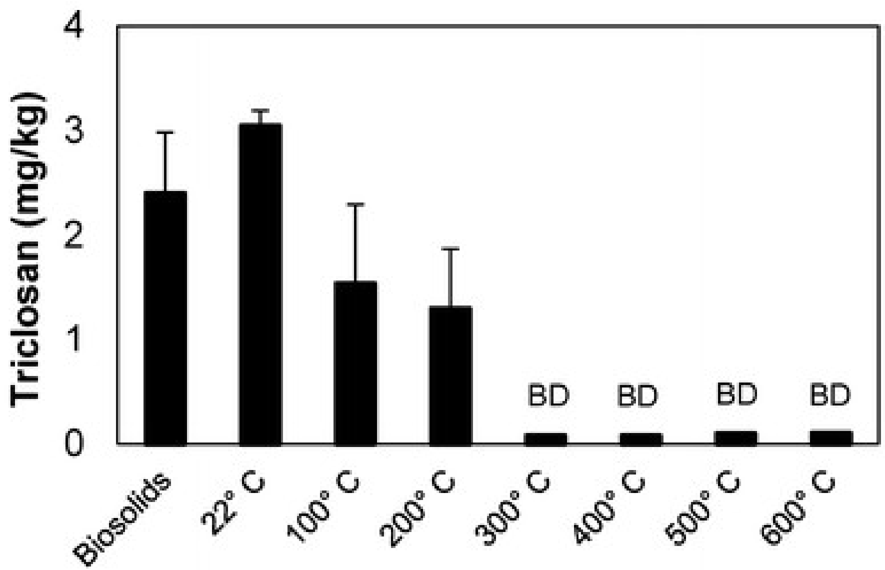
Pyrolysis removes micropollutants from biosolids

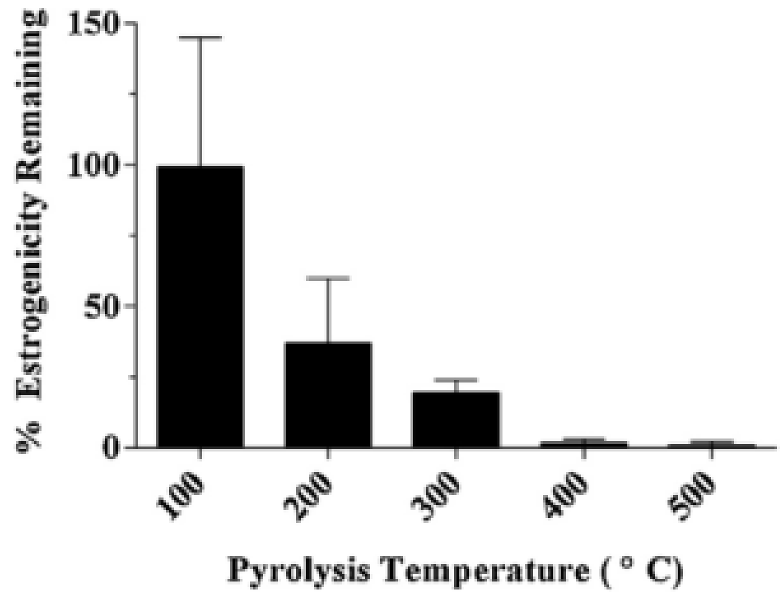


BD: Below Detection Limit

Ross J.J., Zitomer, D.H., Miller, T.R., Weirich, C.A., McNamara, P.J. 2016. Emerging investigators series: pyrolysis removes common microconstituents triclocarban, triclosan, and nonylphenol from biosolids. *Environ. Sci.: Water Res. Technol* **(2)** 282-289.

Pyrolysis removes micropollutants from biosolids





BD: Below Detection Limit

Ross J.J., Zitomer, D.H., Miller, T.R., Weirich, C.A., McNamara, P.J. 2016. Emerging investigators series: pyrolysis removes common microconstituents triclocarban, triclosan, and nonylphenol from biosolids. *Environ. Sci.: Water Res. Technol* **(2)** 282-289.

Hoffman TC., Zitomer, D.J. McNamara, P.J. 2016. Pyrolysis of wastewater biosolids significantly reduces estrogenicity. *J. Haz*. *Mat.* 317, 579-584

Pyrolysis for removal of PFAS

Environmental Science
Water Research & Technology

ROYAL SOCIETY OF CHEMISTRY

PAPER

View Article Online
View Journal

Cite this: DOI: 10.1039/d0ew00763c

Removal of PFASs from biosolids using a semi-pilot scale pyrolysis reactor and the application of biosolids derived biochar for the removal of PFASs from contaminated water†

Sazal Kundu,[a] Savankumar Patel,[a] Pobitra Halder,[a] Tejas Patel,[a] Mojtaba Hedayati Marzbali,[a] Biplob Kumar Pramanik,[b] Jorge Paz-Ferreiro,[a] Cícero Célio de Figueiredo,[c] David Bergmann,[d] Aravind Surapaneni,[de] Mallavarapu Megharaj[fg] and Kalpit Shah[*ae]

Pyrolysis removes PFAS to below detection limit in biochar

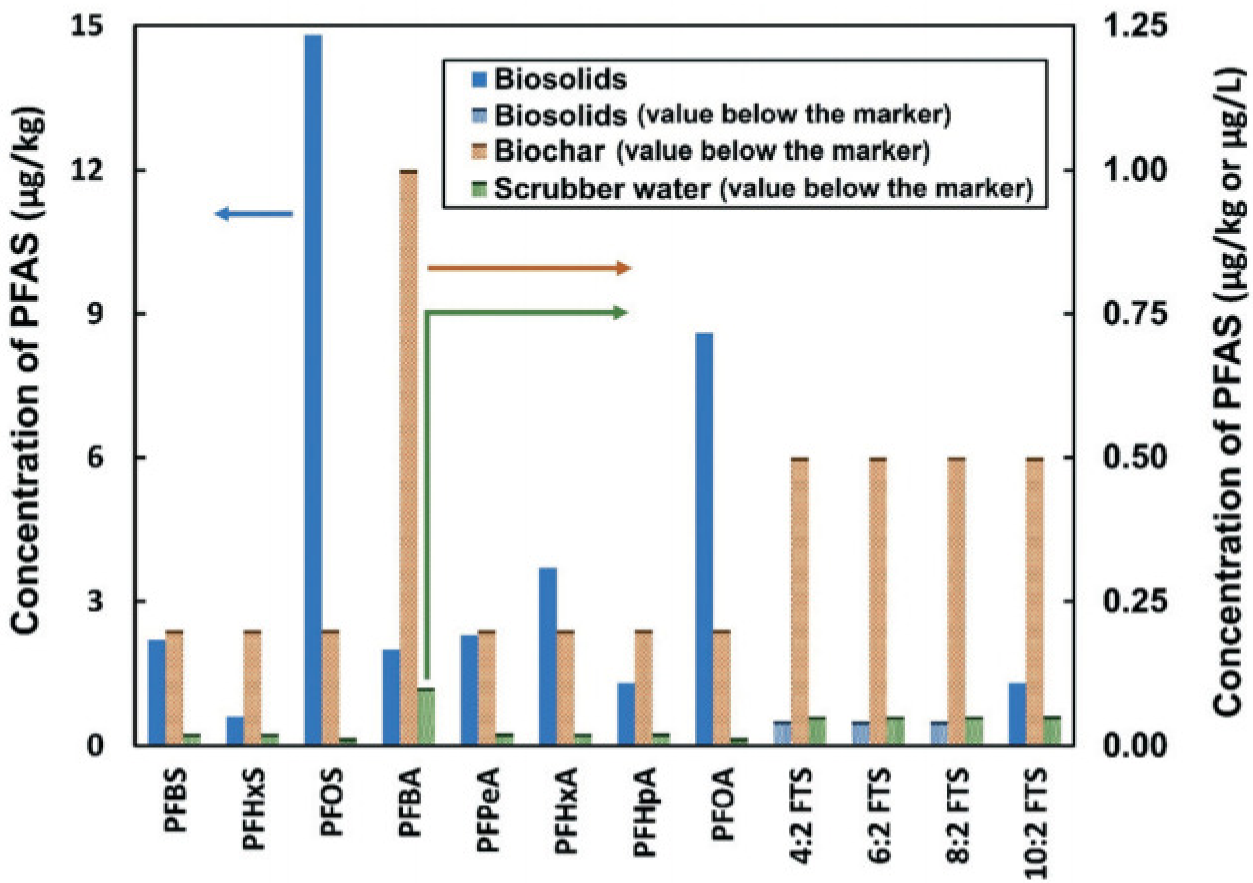


Fig. 8 PFAS concentration data for biosolids (µg kg^{-1}), biochar (µg kg^{-1}) and scrubber water (µg L^{-1}). Columns with markers represent values less than the marker values (see detailed data in Table S1†).

Can changes be made to conventional treatment to improve removal?

The upgrade [from trickling filter to activated sludge] of the WWTF resulted in improved removal efficiency for many endocrine-disrupting chemicals, particularly 17β-estradiol and estrone, and fish exposed to the postupgrade effluent indicated reduction in endocrine disruption relative to preupgrade conditions

Environmental Science & Technology

Article

pubs.acs.org/est

Fish Endocrine Disruption Responses to a Major Wastewater Treatment Facility Upgrade

Larry B. Barber,[*,†] Alan M. Vajda,[‡] Chris Douville,[§] David O. Norris,[||] and Jeffery H. Writer[†]

[†]U.S. Geological Survey, 3215 Marine Street, Boulder, Colorado 80303, United States
[‡]Department of Integrative Biology, University of Colorado, CB 171, Denver, Colorado 80217, United States
[§]City of Boulder, 4049 75th Street, Boulder, Colorado 80301, United States
[||]Department of Integrative Physiology, University of Colorado, 354 UCB, Boulder, Colorado 80309, United States

What if we mitigate discharge?

Results from this whole-lake experiment demonstrate that fish can recover from EE2 exposure at the biochemical through population levels, although the timelines to do so are long for multigenerational exposures. These results suggest that wastewater treatment facilities that reduce discharges of estrogens and their mimics can improve the health of resident fish populations in their receiving environments.

Environmental Science & Technology

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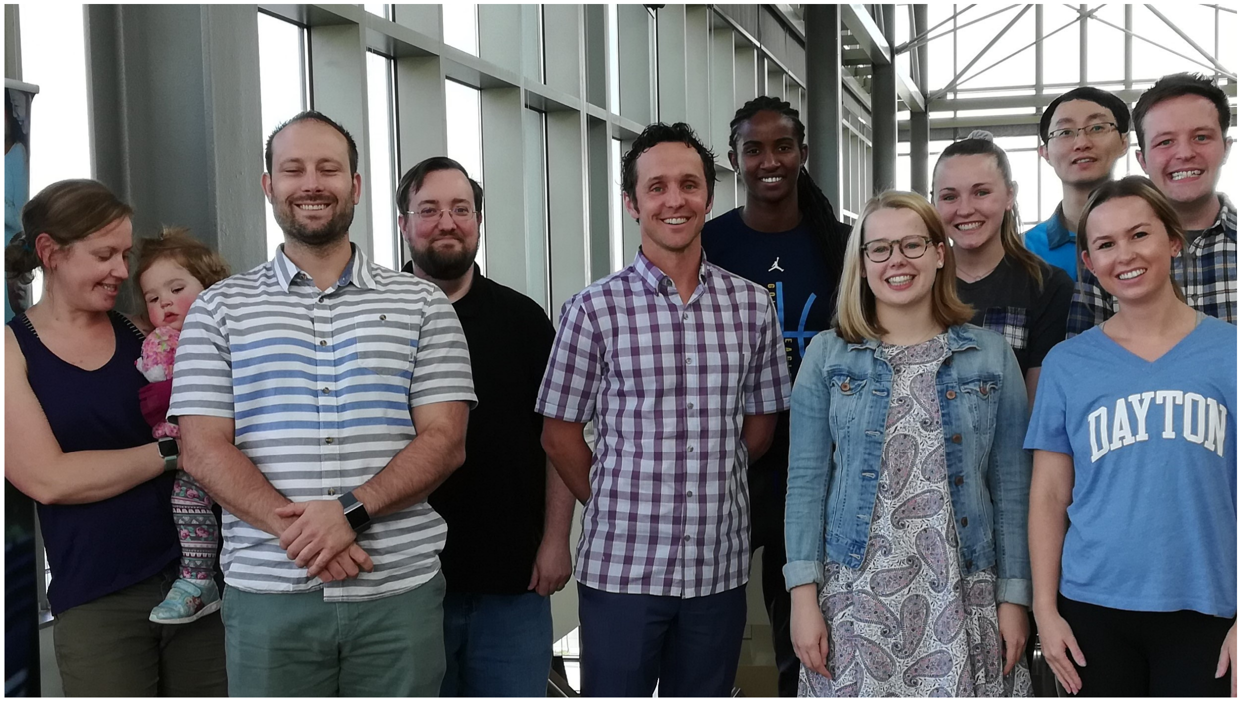
Recovery of a Wild Fish Population from Whole-Lake Additions of a Synthetic Estrogen

Paul J. Blanchfield,[*,†,‡] Karen A. Kidd,[§] Margaret F. Docker,[‡] Vince P. Palace,[†,||] Brad J. Park,[†] and Lianne D. Postma[†]

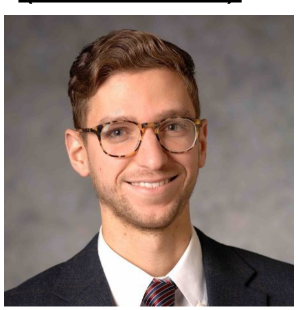
[†]Freshwater Institute, Fisheries and Oceans Canada, 501 University Crescent, Winnipeg, Manitoba R3T 2N6, Canada
[‡]Department of Biological Sciences, University of Manitoba, 50 Sifton Road, Winnipeg, Manitoba R3T 2N2, Canada
[§]Biology Department & Canadian Rivers Institute, University of New Brunswick, Saint John, New Brunswick E2L 4L5, Canada
[||]Stantec Consulting Ltd., 602-386 Broadway Avenue, Winnipeg, Manitoba R3C 3R6, Canada

Acknowledgements

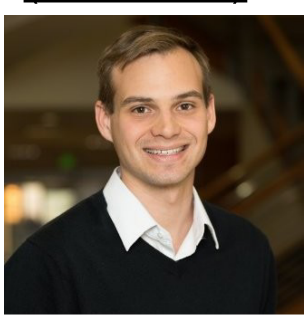


Collaborators
Dr. Zitomer
Dr. Hristova
Dr. Kappell

John Ross (MS 2014)



Tom Hoffman (MS 2015)



Dr. Daniel Carey (PhD 2016)

Dr. Yiran Tong (PhD 2018)

Sponsoring Entities
Lafferty Foundation Grant
Water Quality Center
Marquette University
NSF-WEP Center

Thank you

Questions?

Contact Information:

patrick.mcnamara@marquette.edu

414-288-2188