Pharmaceuticals and Contaminants of Emerging Concern in Domestic Wastewater and their Effects on Septic Systems

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outline

- The world of contaminants
  - Categories & how to prioritize
  - Properties that tell us about their likely removal
- What do we know about occurrence and removal
  - PPCPs and other household compounds, including PFAS
  - Onsite treatment systems
  - Comparison to centralized municipal systems
- Can any of these impact treatment performance?

The Universe of Chemicals

- Elements and their aqueous forms
  - 91 elements with $t_{1/2}$>100 years
  - Each may have as many as 10 isotopes, 11 oxidation states, and many oxo-hydroxyl complexes
- Chemical compounds and ions – most are organic
  - 18.4 M in NIH's PubChem database (9.8 M in Beilstein)
    - ~100,000 new ones each year
  - 800,000 are in active use today
  - 85,000 are or have been readily available in commerce
  - 8,000 currently in high production

At 20 min/compound, lecture ends at 3:50 AM on April 26, 2716

Organic Compounds: Types?

- Natural Compounds
  - Fulvics
  - Proteins, carbohydrates, etc
  - cyanotoxins
- Domestic WW Organics
- Industrial Synthetic Organics
  - Plasticizers: phthalates
  - solvents: tetrachloroethylene
  - waxes: chlorinated parafins
  - others: PCB's
- Hydrocarbons & oil derivatives
  - includes products of combustion: PAH's
- Agricultural Chemicals
  - pesticides: DDT, kepone, mirex

Covered with NOM-Day 1

- Pharmaceuticals, etc
  - Anti-epileptics
  - Beta-blockers
  - X-ray contrast media
  - antibiotics
- Home & Personal Care Products
  - triclosan
  - Musks, flame retardants
- Endocrine Disrupters
  - Steroidal estrogens
- Natural process byproducts
  - Conjugated pharmaceuticals
- Engineered process byproducts
  - disinfection byproducts, etc

Covered with NOM-Day 1 and case studies Day 3

The RISK21 Framework

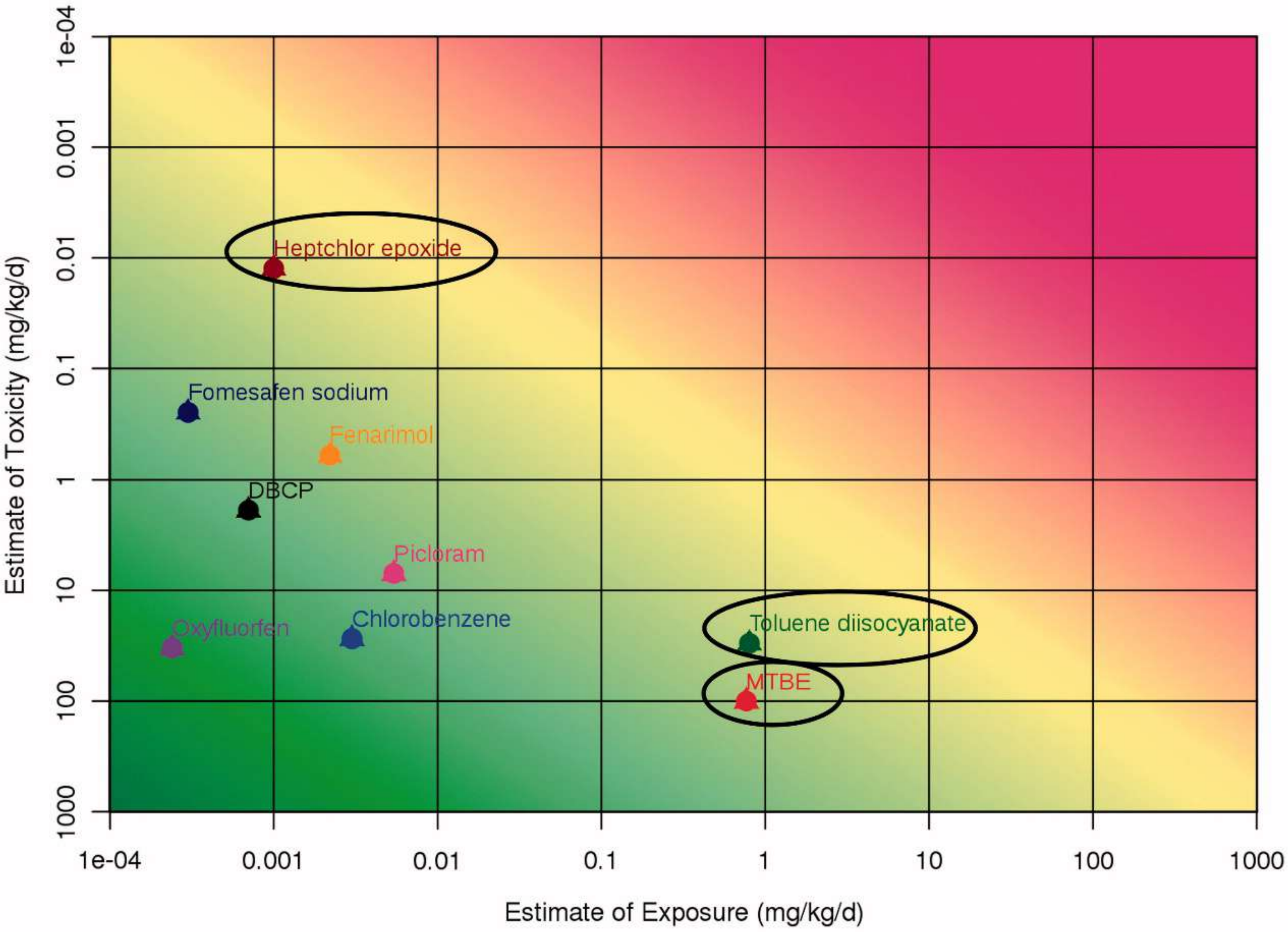


Figure 4. Third evaluation. Matrix plot of the nine remaining chemicals (point estimates for both exposure and toxicity). Those circled are the three designated high priority for further evaluation based on proximity to the yellow zone.

Published in: Douglas C. Wolf; Ammie Bachman; Gordon Barrett; Cheryl Bellin; Jay I. Goodman; Elke Jensen; Angelo Moretto; Tami McMullin; Timothy P. Pastoor; Rita Schoeny; Brian Slezak; Korinna Wend; Michelle R. Embry; *Critical Reviews in Toxicology* **2016,** 46, 43-53.

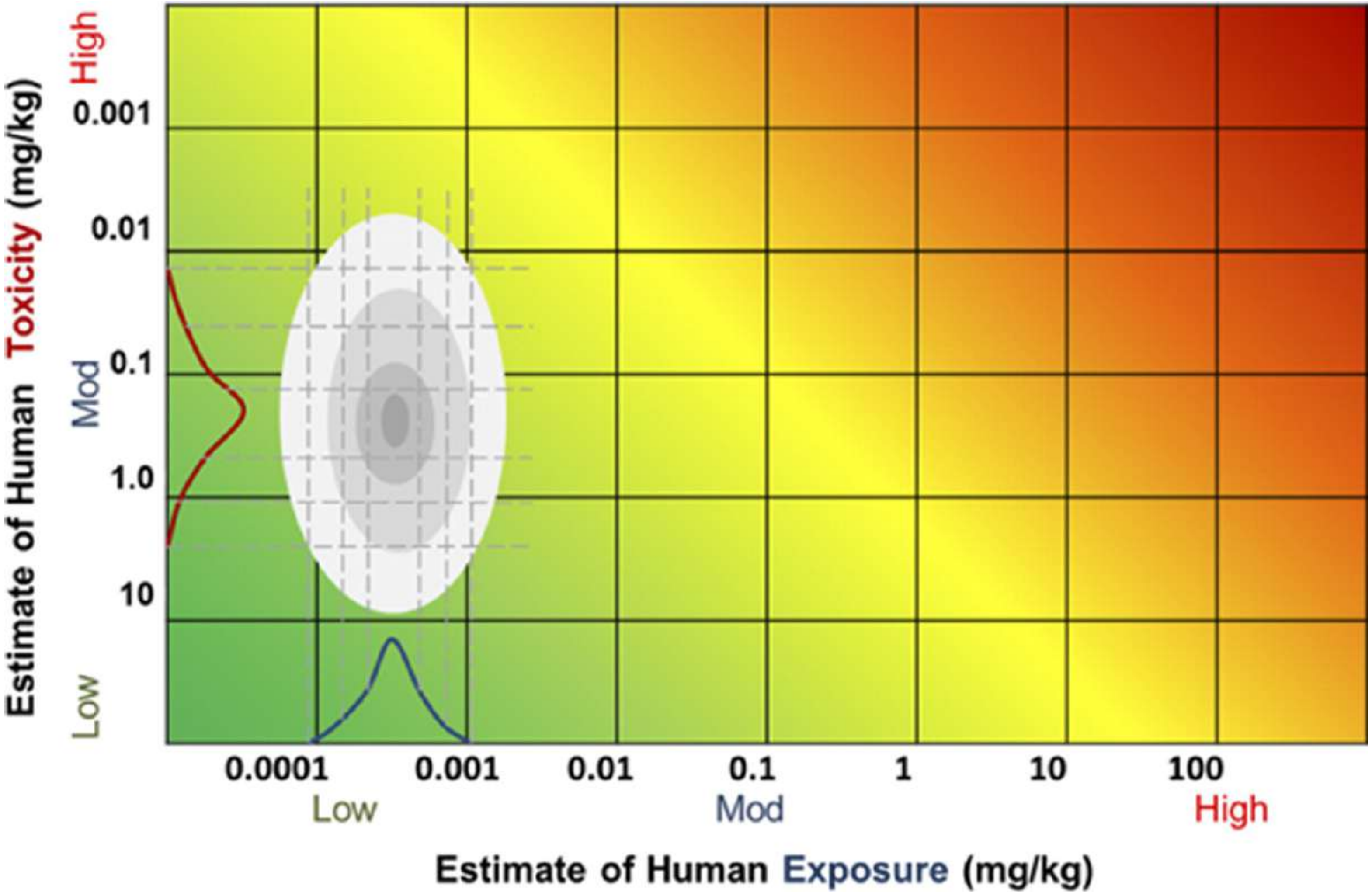
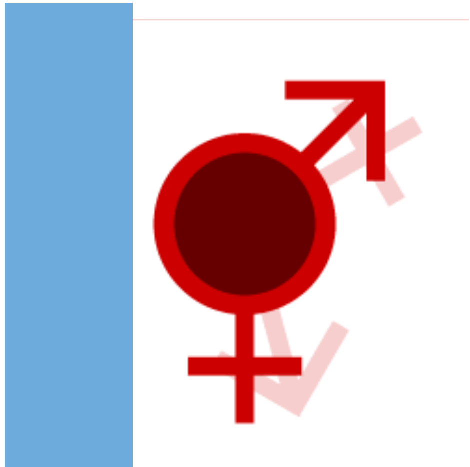


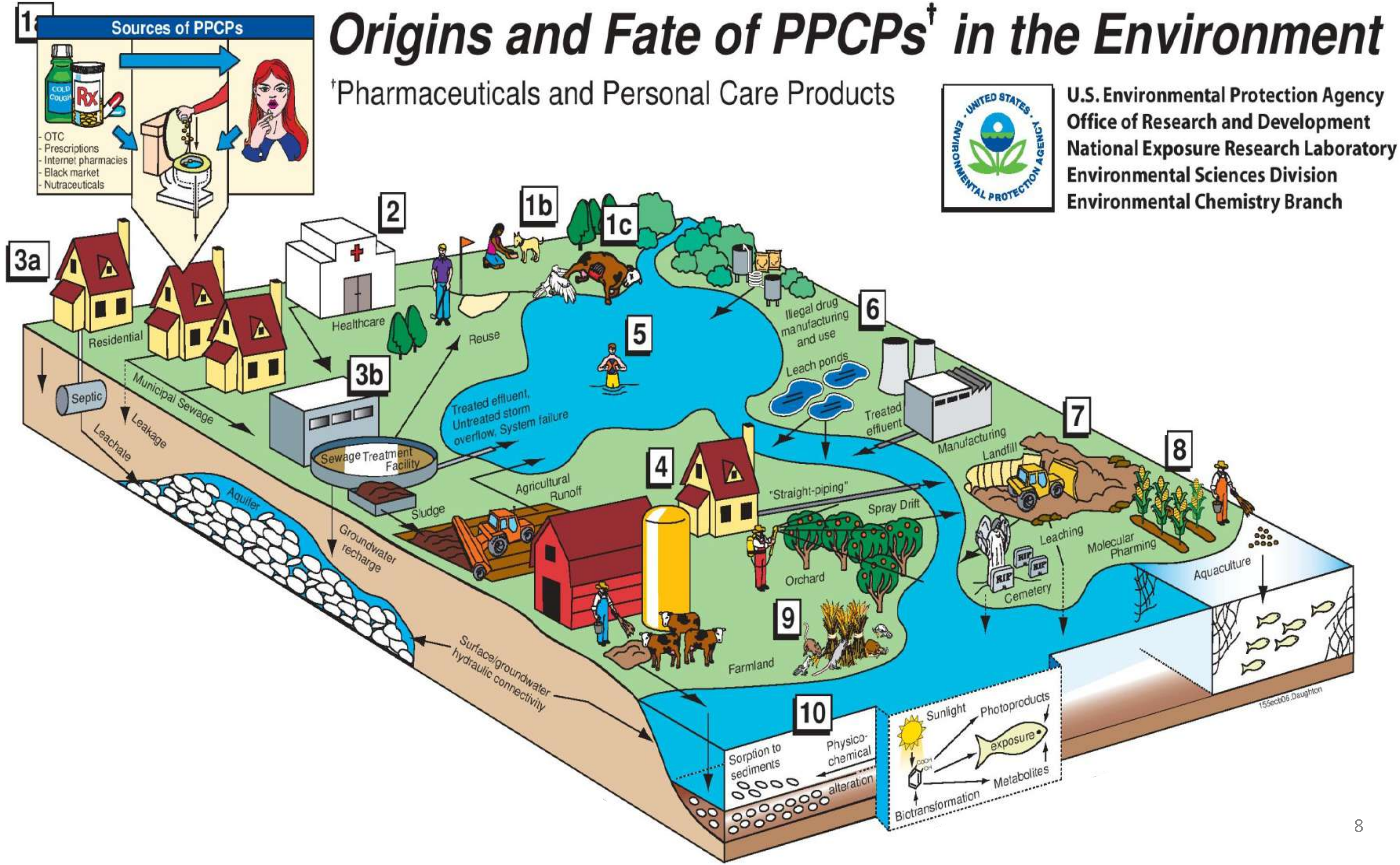
Figure 7. Exposure-toxicity intersection formed from mixing two probability distributions showing isoprob contours.

Published in: Michelle R. Embry; Ammie N. Bachman; David R. Bell; Alan R. Boobis; Samuel M. Cohen; Michael Dellarco; Ian C. Dewhurst; Nancy G. Doerrer; Ronald N. Hines; Angelo Moretto; Timothy P. Pastoor; Richard D. Phillips; J. Craig Rowlands; Jennifer Y. Tanir; Douglas C. Wolf; John E. Doe; *Critical Reviews in Toxicology* **2014,** 44, 6-16.

II. Anthropogenic Substances: EDCs & PPCPs

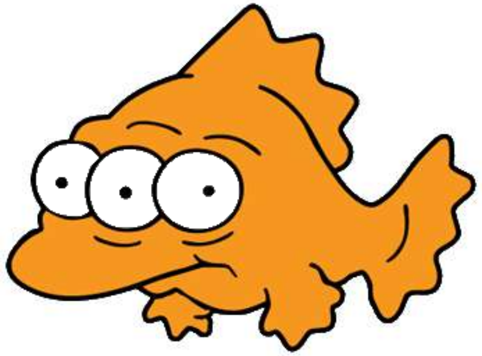
- Endocrine Disrupting Compounds (EDCs)
  - Estrogens: regulate and sustain female sexual development and reproductive function
  - Androgen: male sex hormones
  - Mimics: *estrogenic* and *androgenic* compounds
  - Also anti-estrogenic and anti-androgenic
- Pharmaceuticals and Personal Care Products (PPCPs)
  - Non-steroidal anti-inflammatory
  - Anti-epileptic
  - Antibiotics
  - Anti-anxiety
  - Antioxidants
  - Pain reliever
  - Anti-cholesterol
  - Sun Screen





Origins and Fate of PPCPs† in the Environment
†Pharmaceuticals and Personal Care Products
U.S. Environmental Protection Agency
Office of Research and Development
National Exposure Research Laboratory
Environmental Sciences Division
Environmental Chemistry Branch
1a
Sources of PPCPs
- OTC
- Prescriptions
- Internet pharmacies
- Black market
- Nutraceuticals
1b
1c
2
Healthcare
3a
Residential
Septic
Leachate
Leakage
Municipal Sewage
3b
Sewage Treatment Facility
Sludge
Reuse
Treated effluent, Untreated storm overflow, System failure
Agricultural Runoff
Groundwater recharge
Aquifer
Surface/groundwater hydraulic connectivity
4
5
6
Illegal drug manufacturing and use
Leach ponds
Treated effluent
Manufacturing
7
Landfill
Leaching
Cemetery
8
Molecular Pharming
Aquaculture
9
Farmland
Orchard
"Straight-piping"
Spray Drift
10
Sorption to sediments
Physico-chemical alteration
Sunlight
Photoproducts
exposure
Metabolites
Biotransformation
155ecb06.Daughton

What are OWCs, EDCs and PPCPs and Why the Interest?

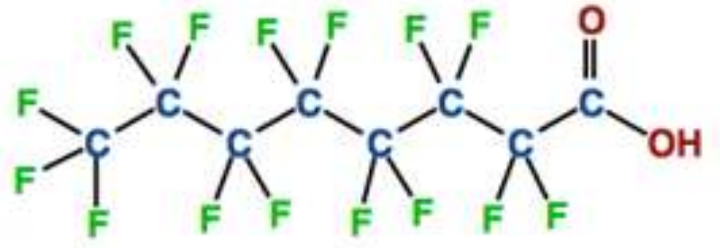


- **EDC**- Endocrine Disrupting Compounds
  - EDCs are a class of compounds which alter
    the hormonal system of an organism.
  - Eg: DDT, 17-alpha Ethinylestradiol, Bisphenol A,etc.
- **PPCP**- Pharmaceuticals and Personal Care Products
  - Any products used for personal health or cosmetic reasons
  - Includes prescription and non-prescription drugs, veterinary drugs, fragrances and cosmetics
- **OWC** – Organic wastewater compounds
  - All of the above and more

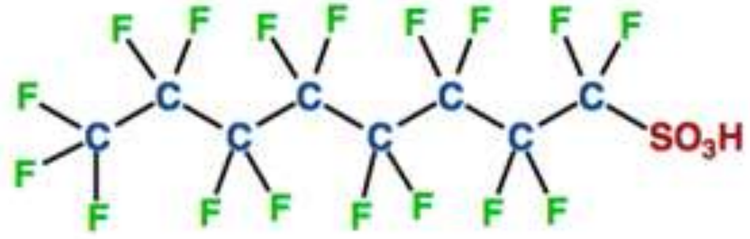
- Why study them?
  - Direct impacts on human health
    - Maybe not the most important?
    - Impacts of mixture are uncertain
  - Public perception
    - Becoming a very sensitive issue
  - Direct impacts on ecological health
    - Well documented: feminization of fish, etc.
  - Tracers of wastewater contamination
  - Indicators & promoters of antibiotic resistance
  - Precursors to more Hazardous DBPs

<u>Fluorinated hydrocarbons</u>: nomenclature

- Poly- and Perfluoroalkyl substances (PFAS)
    - **<u>Per</u>** means all hydrogens are substituted with fluorine atoms
    - **<u>Poly</u>** means more than one fluorine atom, but some hydrogens too
- Perfluoroalkyl acids (PFAAs)
    - Perfluorocarboxylic Acids
        - C4 to C12 compounds measured
        - C8 was in CCL3: PFOA
    - Perfluorosulfonic Acids
        - C4 to C10 compounds measured
        - C8 was in CCL3: PFOS
    - Many others, e.g.,
        - Perflurosulfonamides
        - Perflorosulfonamidoacetic acids



PFOA - perfluorooctanoic acid



PFOS - perfluorooctanesulfonic acid

More on PFAS at the end

Estimating Source Terms

- Use-based calculations (e.g., Sedlak)
  - Get national or regional use data
  - Estimate non-metabolized/adsorbed fraction
  - Estimate removal across conventional WWT
- Real WW effluent monitoring
  - Highly variable based on date, time, location, processes, climate, etc

Sui et al., 2011

Environmental Science & Technology
ARTICLE
pubs.acs.org/est

Seasonal Variation in the Occurrence and Removal of Pharmaceuticals and Personal Care Products in Different Biological Wastewater Treatment Processes

Qian Sui, Jun Huang, Shubo Deng, Weiwei Chen, and Gang Yu*

School of Environment, THU – VEOLIA Joint Research Center for Advanced Environmental Technology, Tsinghua University, Beijing 100084, China

Supporting Information

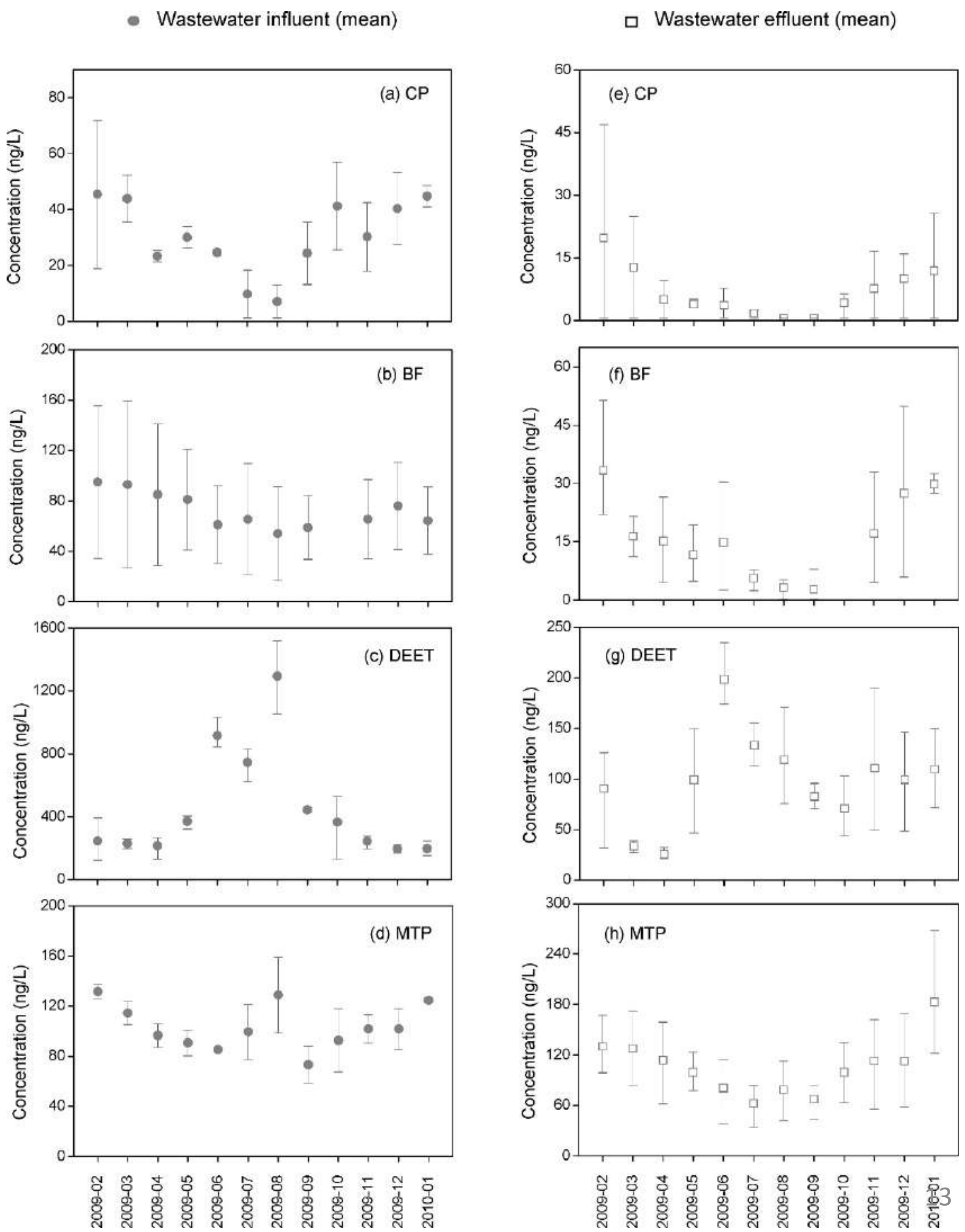
ABSTRACT: The occurrence of 12 pharmaceuticals and personal care products (PPCPs) in two wastewater treatment plants in Beijing was studied monthly over the course of one year. The removal of PPCPs by three biological treatment processes including conventional activated sludge (CAS), biological nutrient removal (BNR), and membrane bioreactor (MBR) was compared during different seasons. Seasonal variations of PPCPs in the wastewater influent were discrepant, while in the wastewater effluent, most PPCPs had lower concentrations in the summer than in the winter. For the easily biodegradable PPCPs, the performance of MBR was demonstrated to be more stable than CAS or BNR especially during winter months. Diclofenac, trimethoprim, metoprolol, and gemfibrozil could be moderately removed by MBR, while their removal by CAS and BNR was much lower or even negligible. Nevertheless, no removal was achieved regardless of the season or the treatment processes for the recalcitrant PPCPs. Studies on the contribution of each tank of the MBR process to the total removal of four biodegradable PPCPs indicated the oxic tank was the most important unit, whereas membrane filtration made a negligible contribution to their elimination.

Sui, Q., Huang, J., Deng, S.B., Chen, W.W. and Yu, G. (2011) Seasonal Variation in the Occurrence and Removal of Pharmaceuticals and Personal Care Products in Different Biological Wastewater Treatment Processes. Environmental Science & Technology 45(8), 3341-3348.

Seasonal Variability

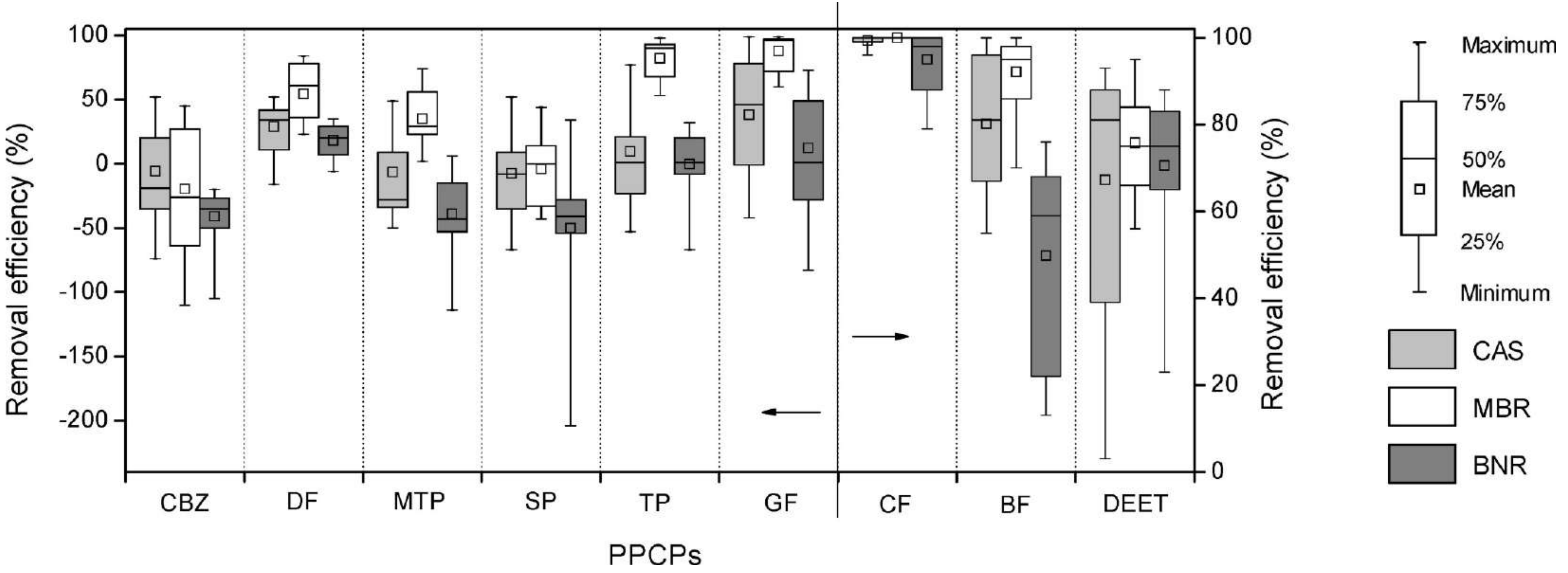
- **BF** → Bezafibrate
- **CBZ** → Carbamazapine
- **CF** → Caffeine
- **CP** → Chloramphenicol
- **DEET** → N,N-diethyl-m-toluamide
- **DF** → Diclofenac
- **GF** → Gemfibrozil
- **MTP** → Metoprolol
- **SP** → Sulpiride
- **TP** → Trimethoprim

Seasonal variation in the concentrations of some PPCPs in the wastewater influents (a–d) and effluents (e–h). The symbols represent the mean concentration, and error bars represent the maximum and minimum concentration in CAS, MBR, and BNR processes.



Qian Sui; Jun Huang; Shubo Deng; Weiwei Chen; Gang Yu; *Environ. Sci. Technol.* 2011, 45, 3341-3348.

Process Removal Efficiencies

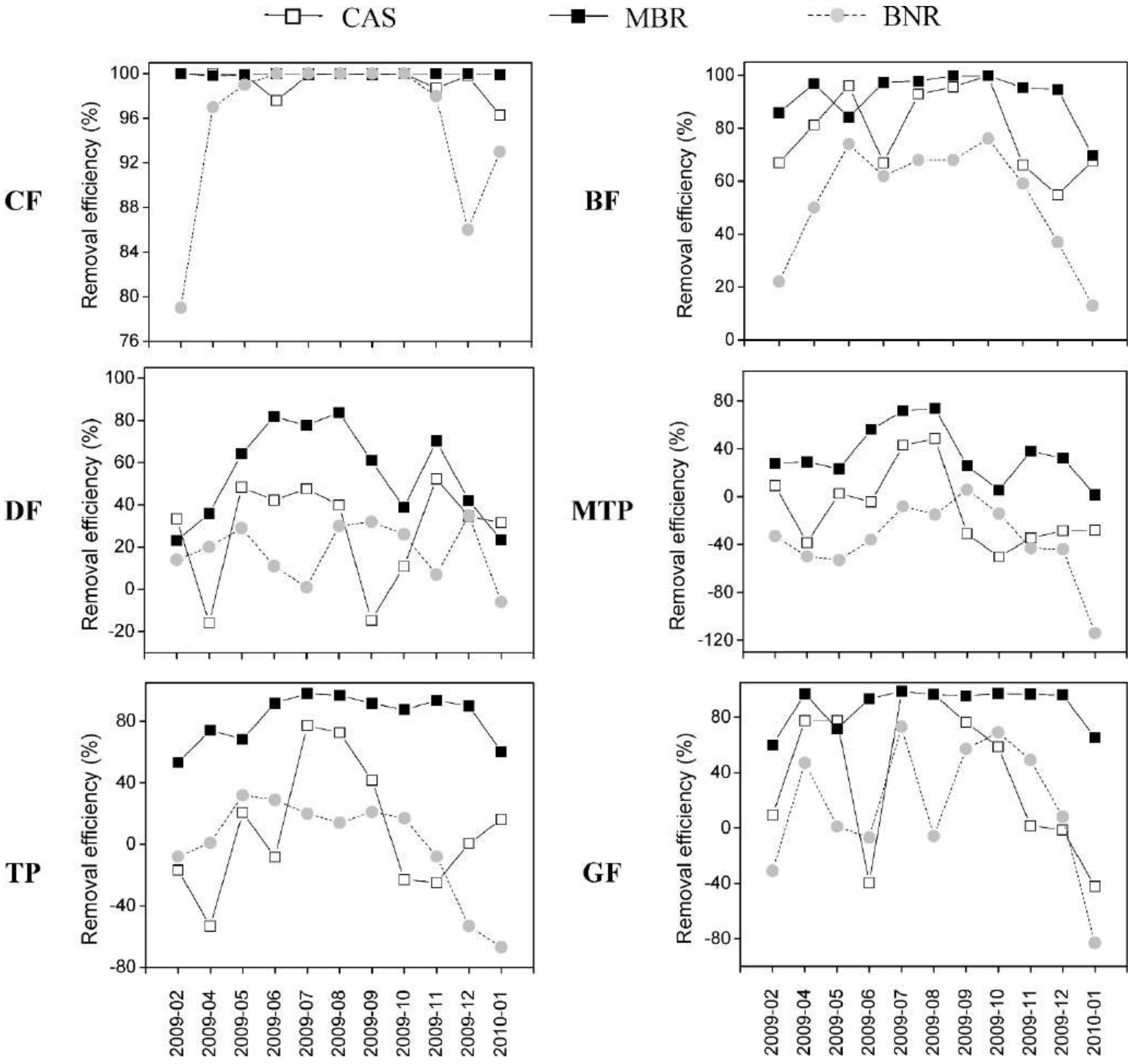


Comparison of the overall removal efficiencies by:

- Conventional Activated Sludge (CAS)
- Biological Nutrient Removal (BNR), and
- Membrane Bioreactor (MBR) processes.

Qian Sui; Jun Huang; Shubo Deng; Weiwei Chen; Gang Yu;
Environ. Sci. Technol. **2011, 45, 3341-3348.**

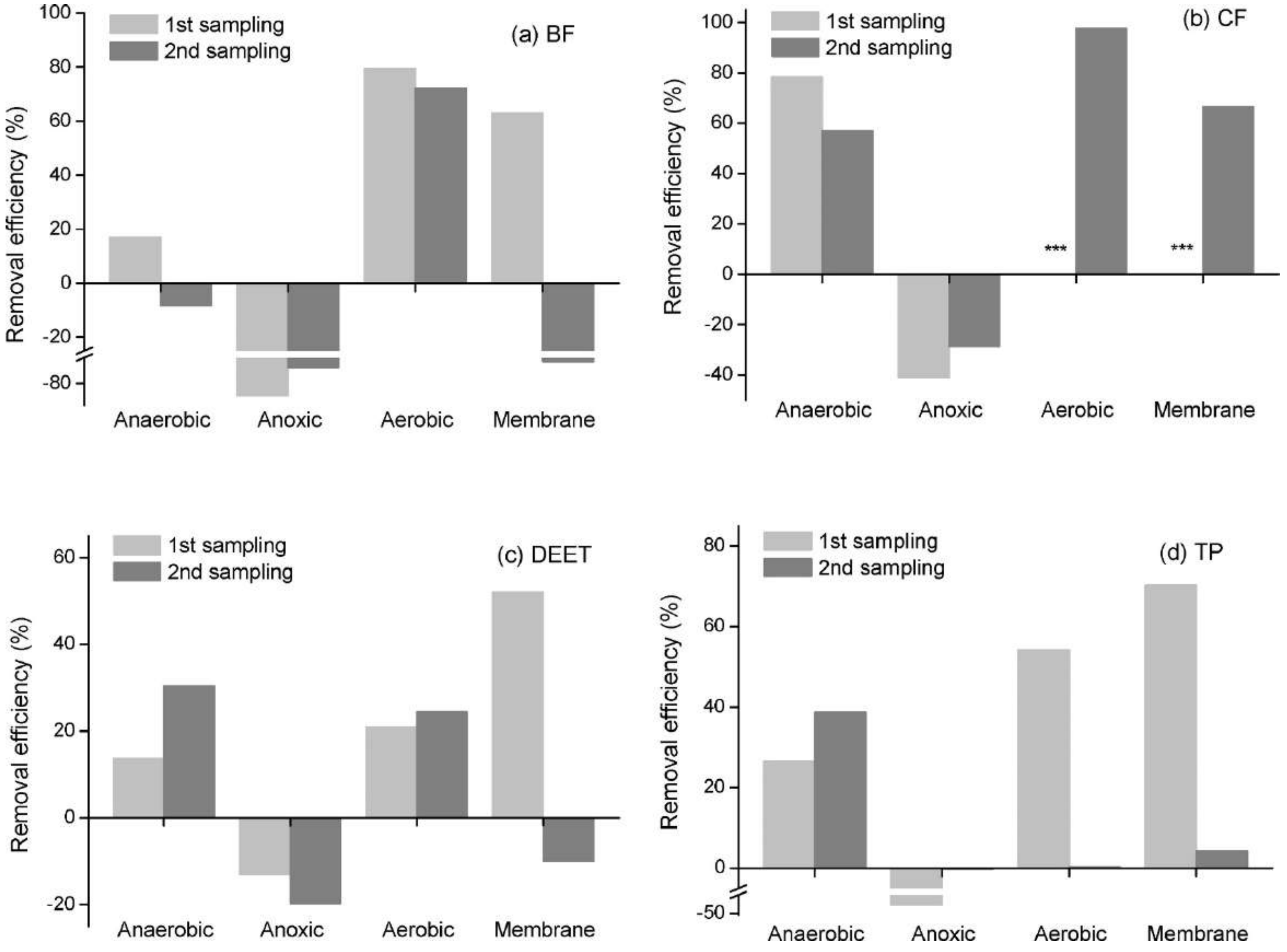
Seasonal variability for removal



Seasonal variation in the removal efficiencies of PPCPs during the whole year: comparison among MBR and other two biological treatment processes.

Qian Sui; Jun Huang; Shubo Deng; Weiwei Chen; Gang Yu; *Environ. Sci. Technol.* 2011, 45, 3341-3348.

Removal across MBR Process



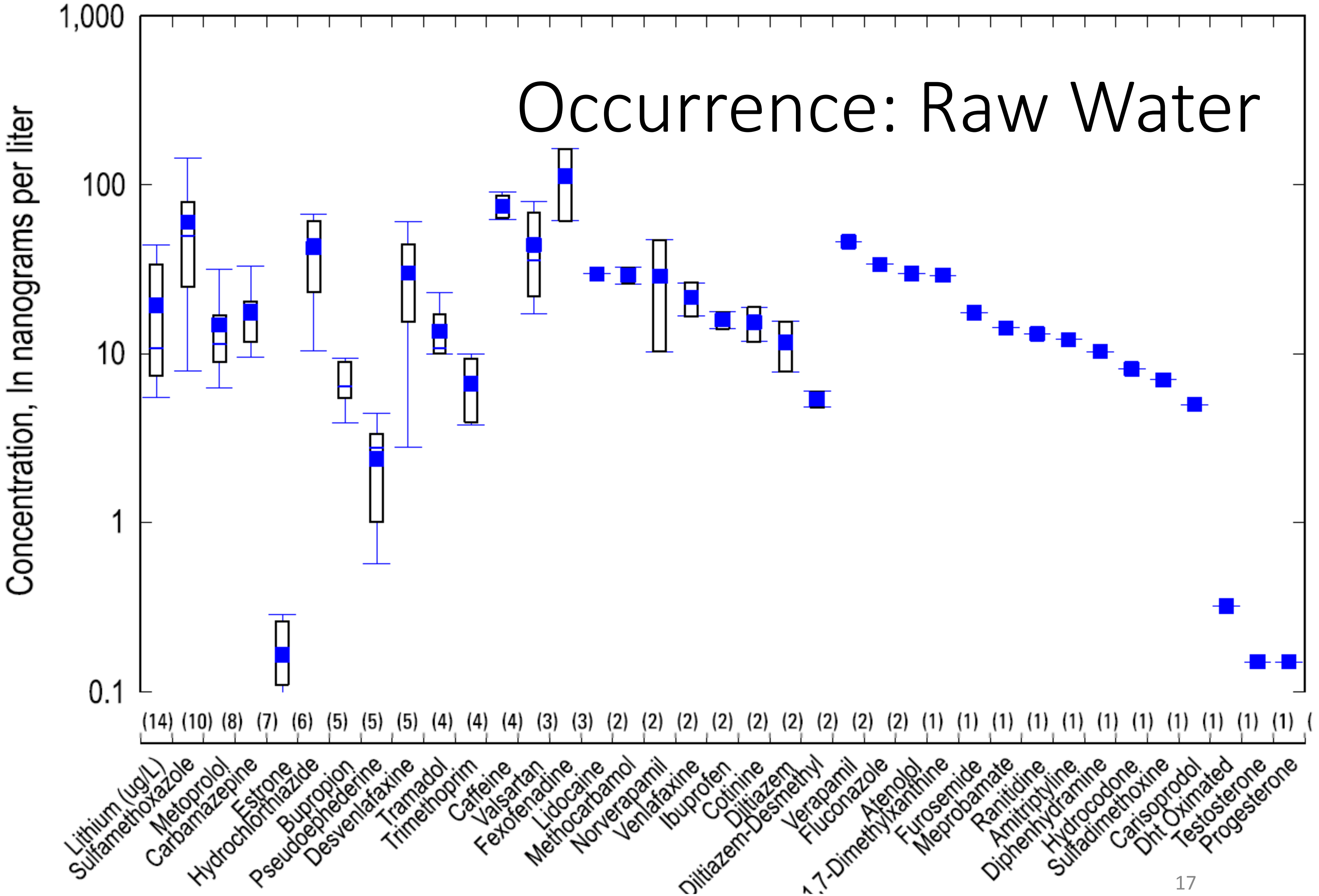
Removal efficiencies of PPCPs in each tank of A/A/O-MBR process: (a) BF, (b) CF, (c) DEET, (d) TP. *** means that the removal efficiency of aerobic tank and membrane filtration could not be calculated because the CF concentrations were <LOQ after anoxic tank in the first sampling.

Qian Sui; Jun Huang; Shubo Deng; Weiwei Chen; Gang Yu;
Environ. Sci. Technol. 2011, 45, 3341-3348.

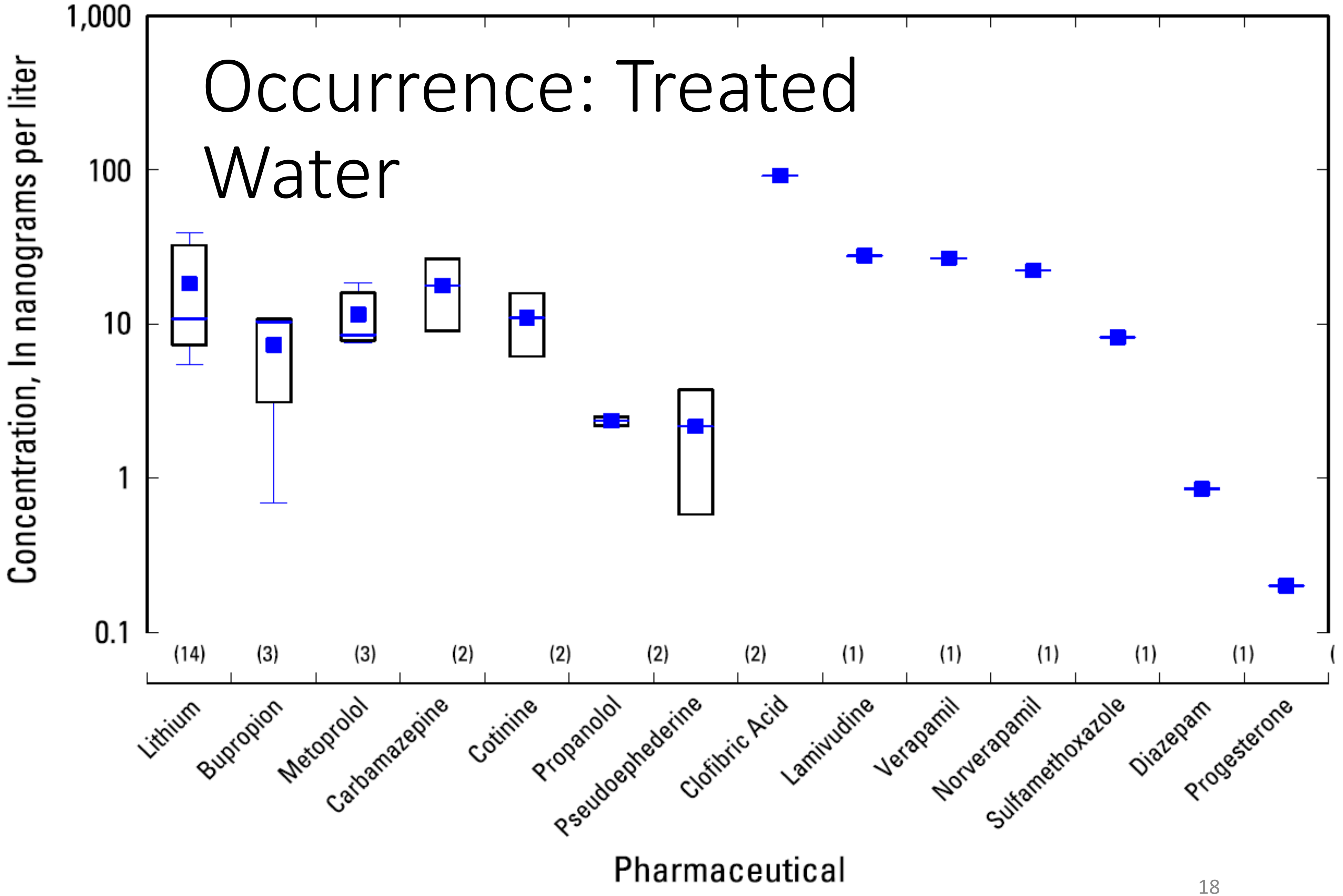
Occurrence: Raw Water

- 25 systems monitored

From: Glassmeyer, 2016



Occurrence: Treated Water



From: Glassmeyer, 2016

Generic Mechanistic View

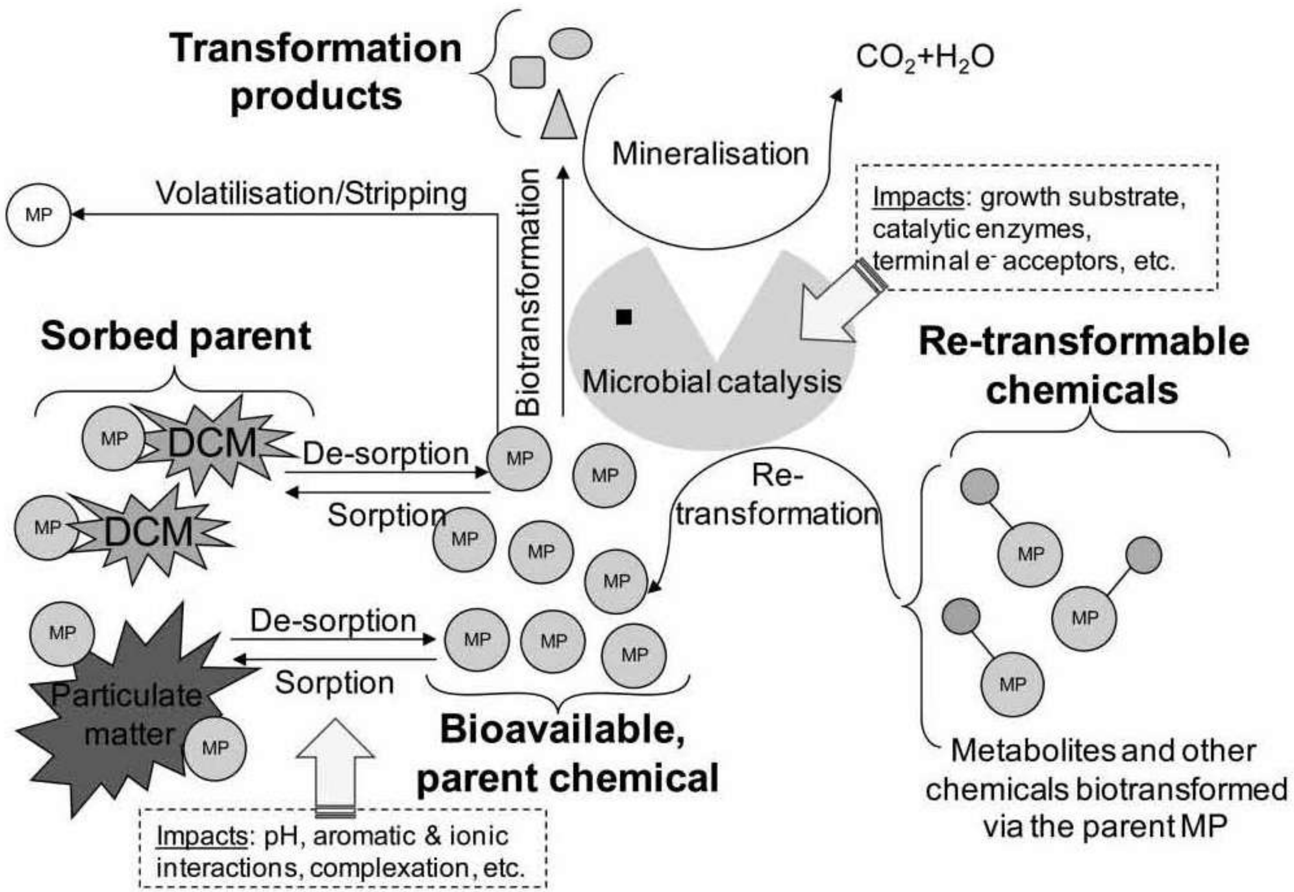


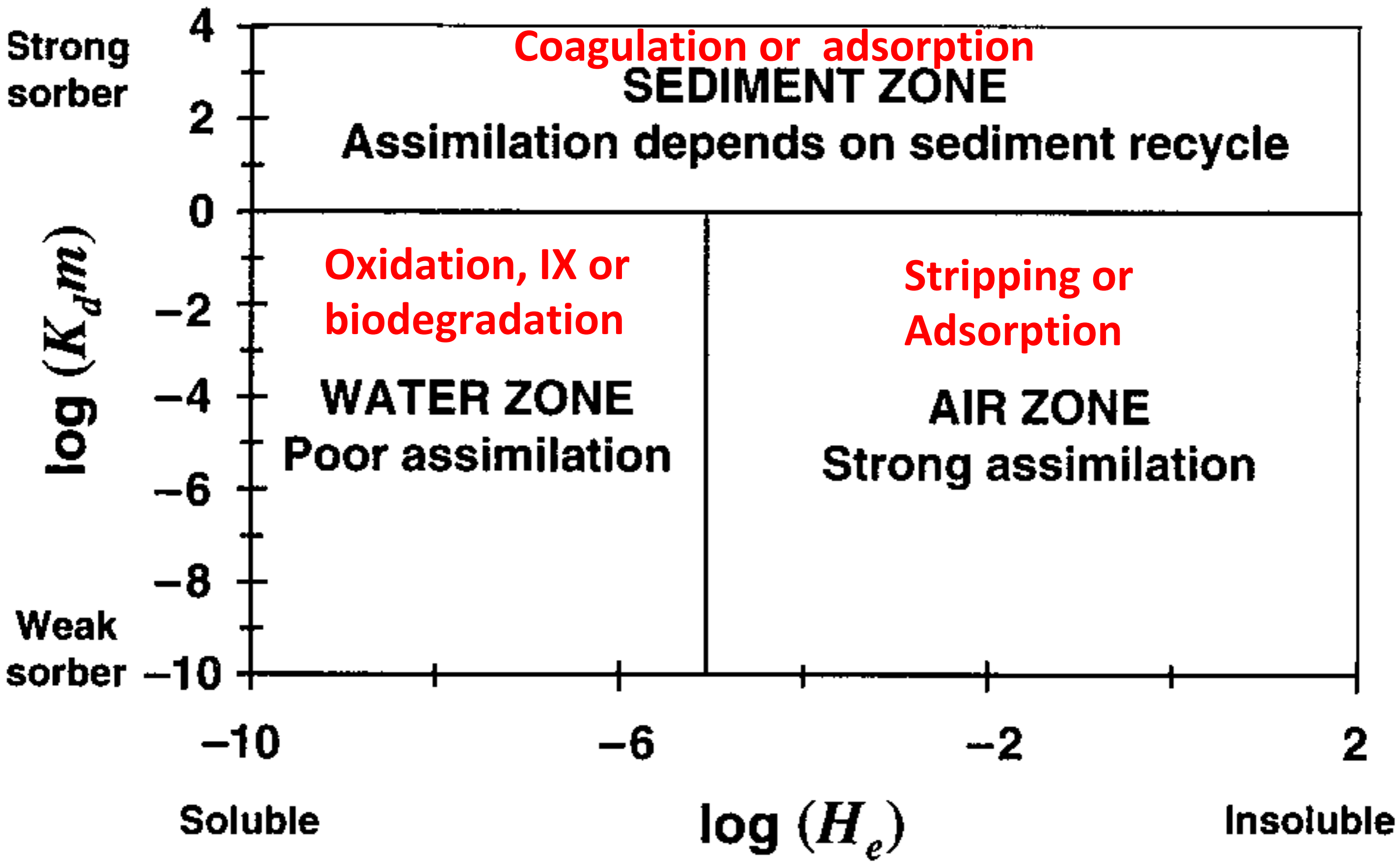
Figure 1 | Micro-pollutant (MP) fractions and processes, influencing MP removal in wastewater. Compiled based on studies by Criddle (1993); Alvarez-Cohen & Speitel (2001); Ternes & Joss (2006); Melcer *et al.* (2007); Monteith *et al.* (2008); Lindblom *et al.* (2009); Barret *et al.* (2010a); Plósz *et al.* (2010b,c). DCM: dissolved and colloidal matter.

Plosz, B.G., Benedetti, L., Daigger, G.T., Langford, K.H., Larsen, H.F., Monteith, H., Ort, C., Seth, R., Steyer, J.P. and Vanrolleghem, P.A. (2013) Modelling micro-pollutant fate in wastewater collection and treatment systems: status and challenges. Water Science and Technology 67(1), 1-15.

Fate and Transport Modeling

Chemical Properties => Destiny

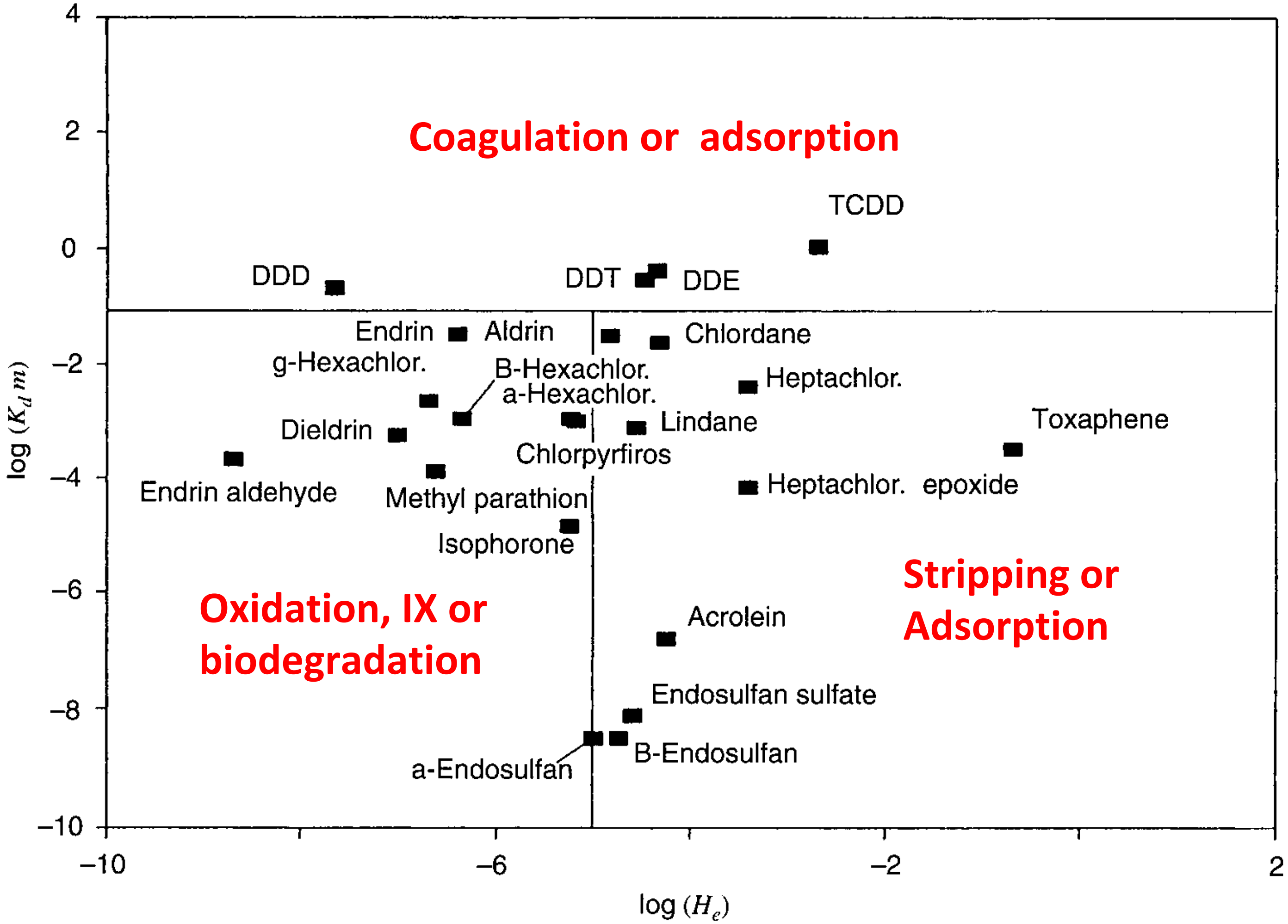
Chapra, 1991 [ASCE JEED 17(5)656]



- Fate in Aquatic systems
  - Relates to fate in treatment
- Aquatic assessment assuming:
  - T_a=283 K
  - M=200 g/mole
  - U_w = 5 mph
  - v_s =91 m/yr
- Assimilation refers to general rate of removal

Summary: pesticides

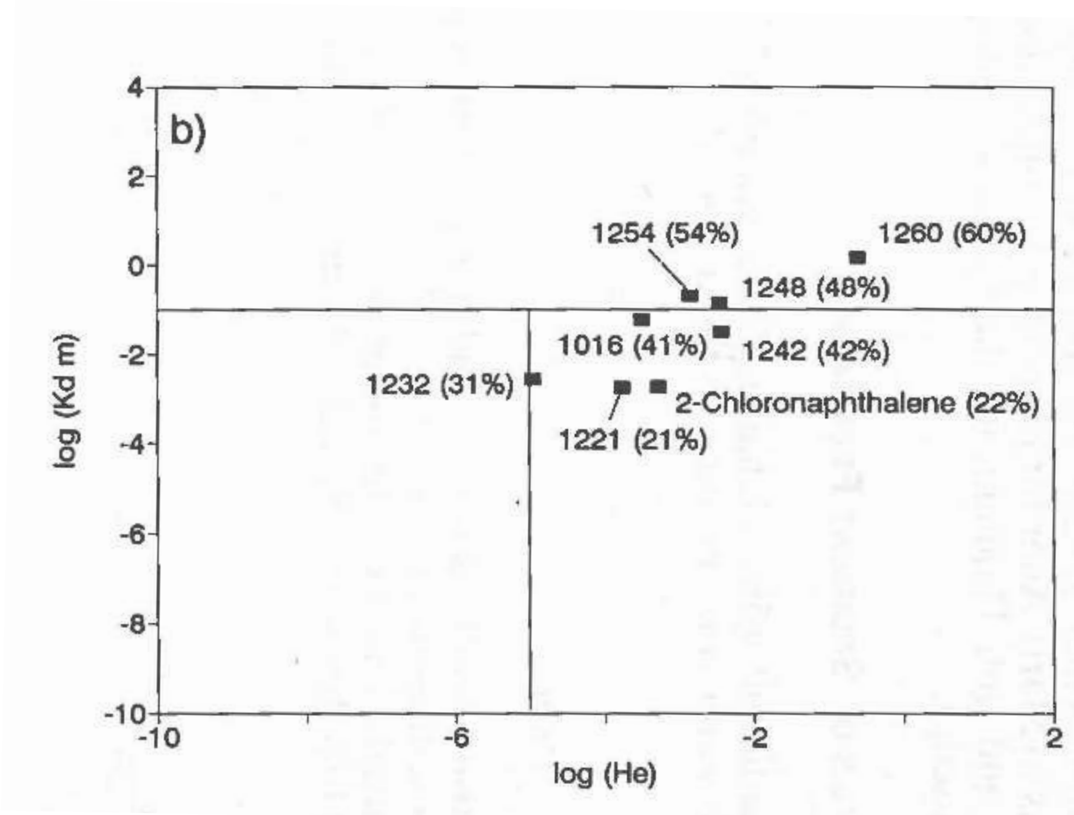
- Modified from: Surface Water Quality Modeling, by Chapra, pg.735



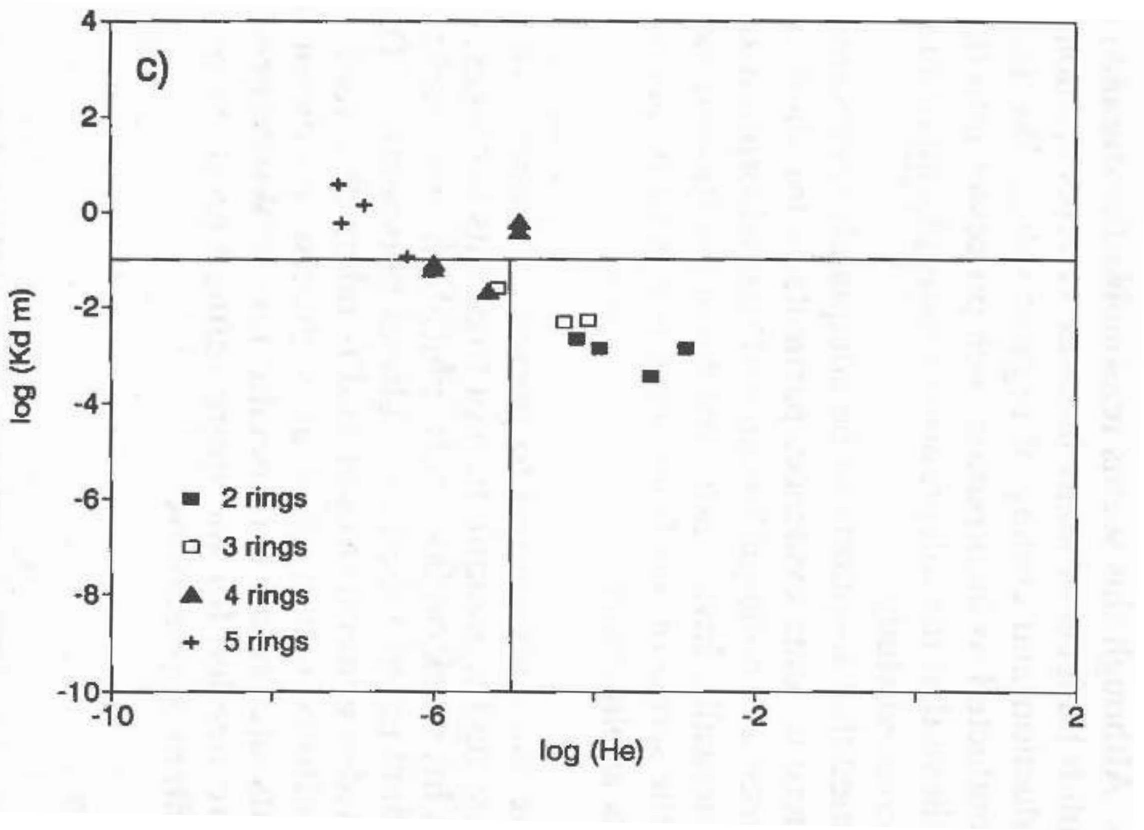
Chapra, 1991 [ASCE JEED 17(5)656]

Aromatic Carcinogens

- PCBs (Polychlorinated Biphenyls)
  - Arochlor mixtures



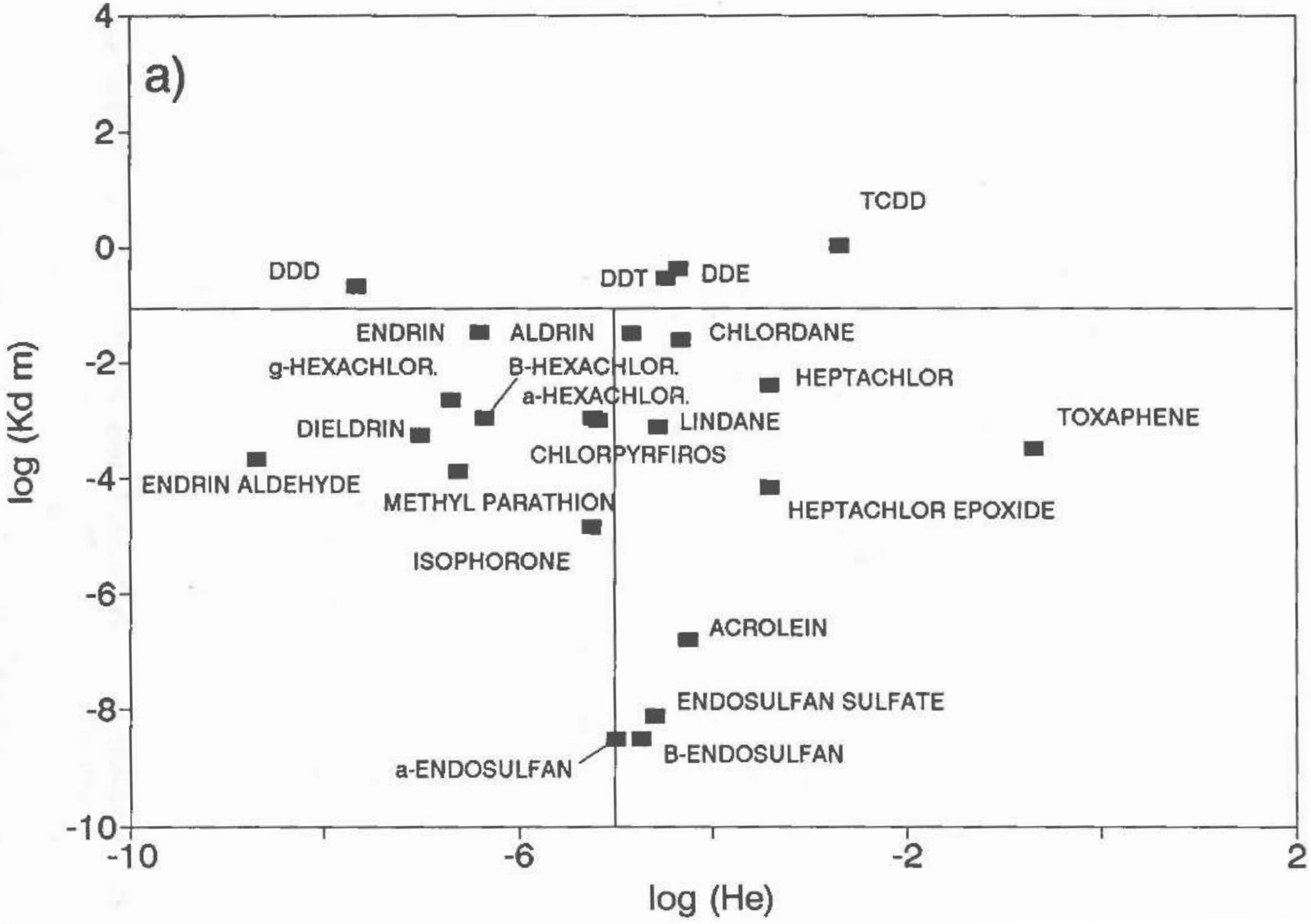
- PAHs (polynuclear aromatic hydrocarbons)



Chapra, 1991 [ASCE JEED 17(5)656]

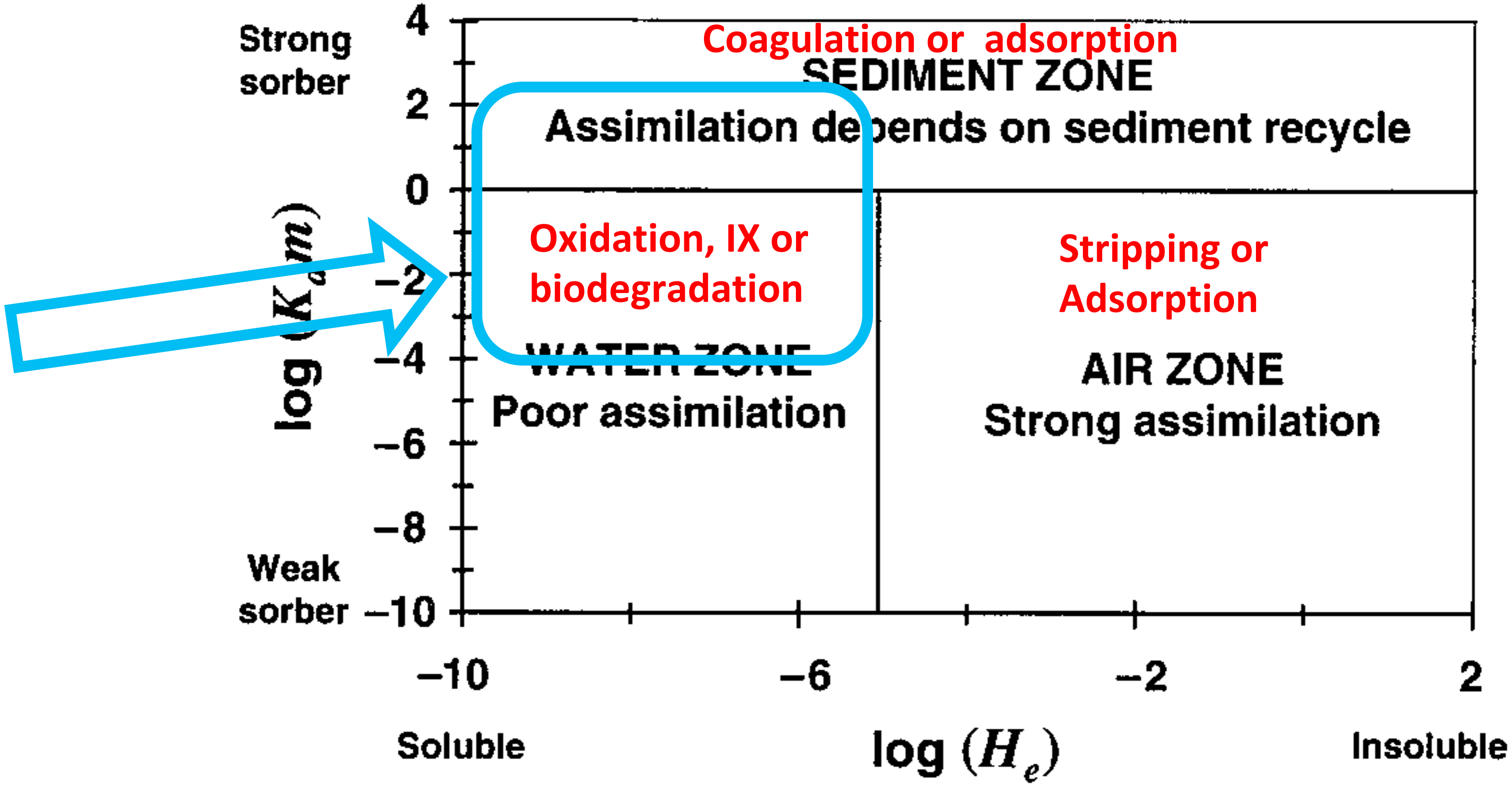
Pesticides

- Broad spectrum of properties



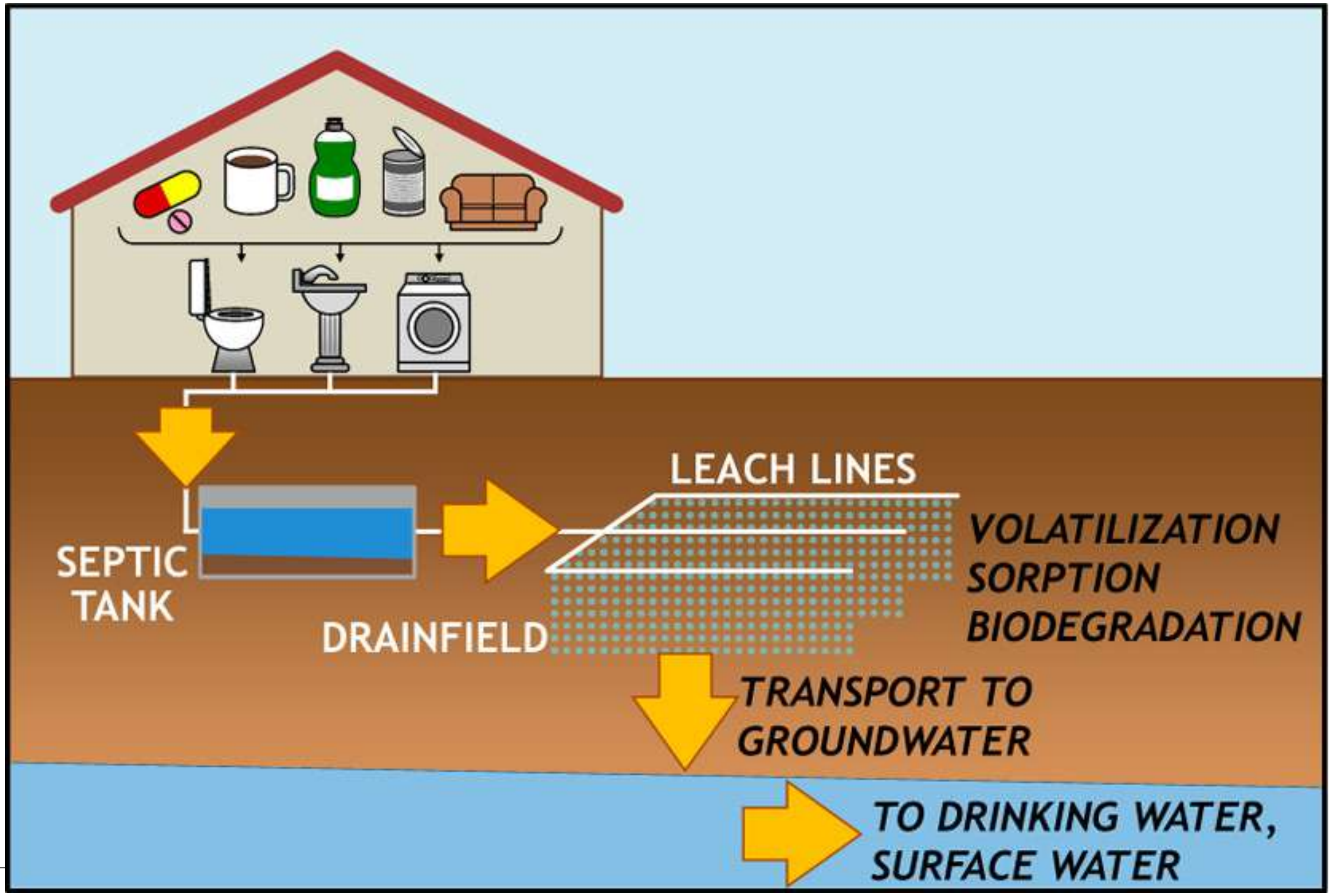
Where are the PPCPs and OWCs?

- Not much data on volatilization, but we do have K_{ow} values
- Most are probably in this region



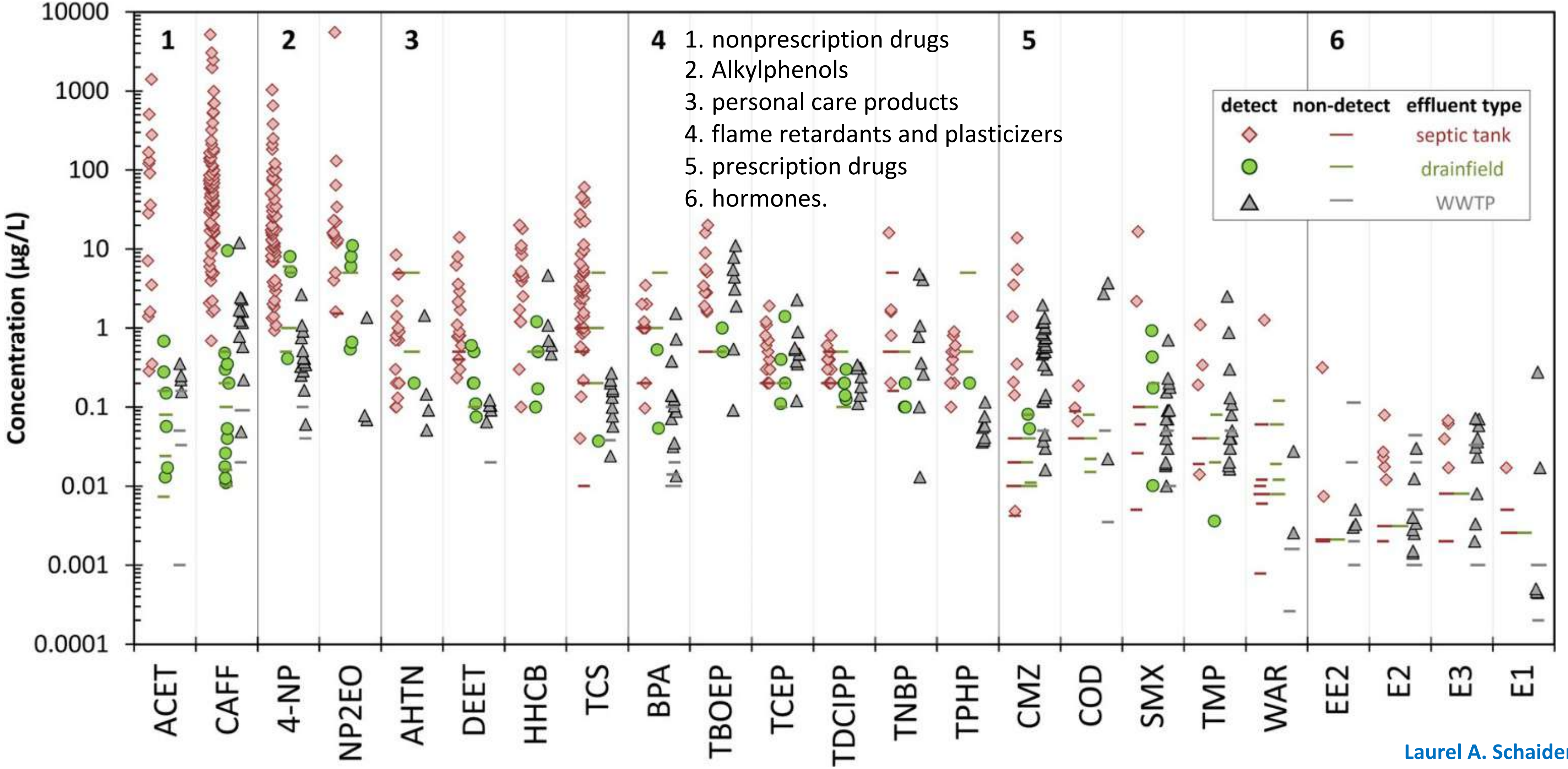
Onsite Wastewater Treatment Systems

- Conventional
  - Septic Tank
  - Drainfield
- Alternative
  - Biofilters
  - Aerobic units
  - Special sorbents



Laurel A. Schaider; Kathryn M. Rodgers; Ruthann A. Rudel; *Environ. Sci. Technol.* 2017, 51, 7304-7317.

Effluent Organic Wastewater Contaminants (OWCs)



Concentrations of OWCs in septic tank effluent, drainfield effluent, and WWTP effluent. Horizontal lines show censoring values for systems where the median value was below the censoring value.

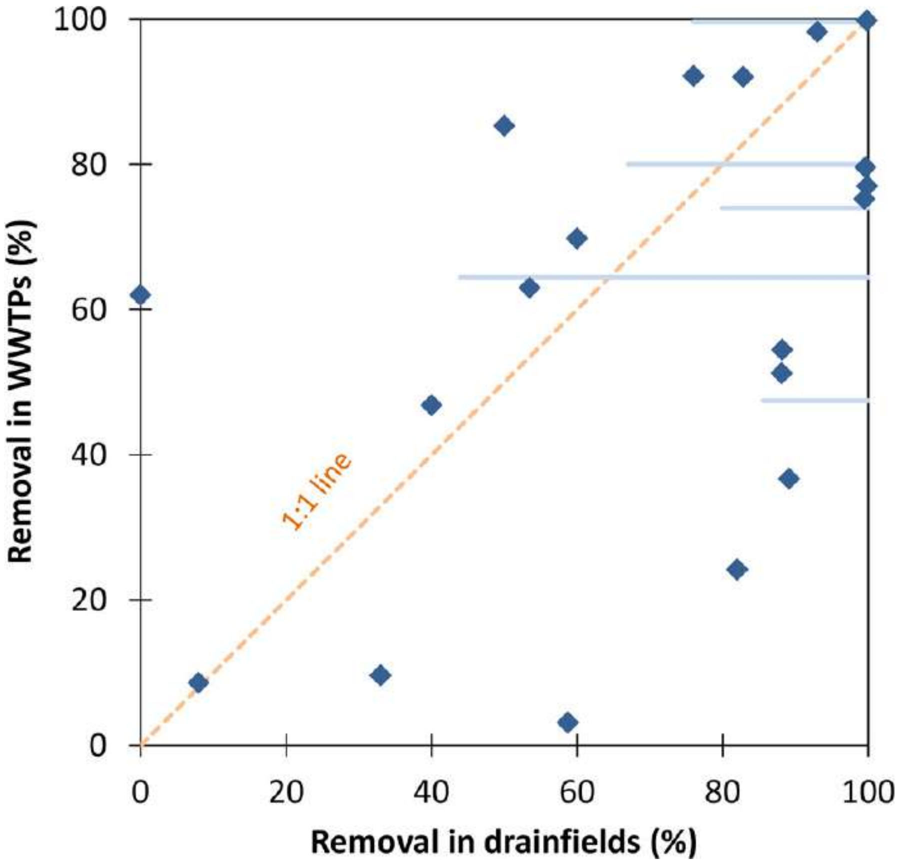
Laurel A. Schaider; Kathryn M. Rodgers; Ruthann A. Rudel; *Environ. Sci. Technol.* 2017, 51, 7304-7317.

Removals of OWCs

- Septic tank
  - Effluent similar to primary effluent in a conventional wastewater treatment plant
  - Mostly due to association to solids, grease, some anaerobic degradation
- Drainfields
  - Removal by sorption, aerobic biodegradation, and some volatilization
  - Some are very well removed (>99%)
    - Triclosan, Caffeine, acetaminophen
      - Presence of these indicates a failed septic system
  - Some are not removed at all
    - Example: certain artificial sweeteners, especially sucralose
      - Presence of these indicates zone of influence

Removal in Drainfields

- Drainfields viewed as analogous to activated sludge systems
- Median effluent concentrations are similar
  - Vary by < factor of 10 for 24 of 29 OWCs
  - A few were much lower in drainfield effluents
    - Trimethoprim (antibiotic)



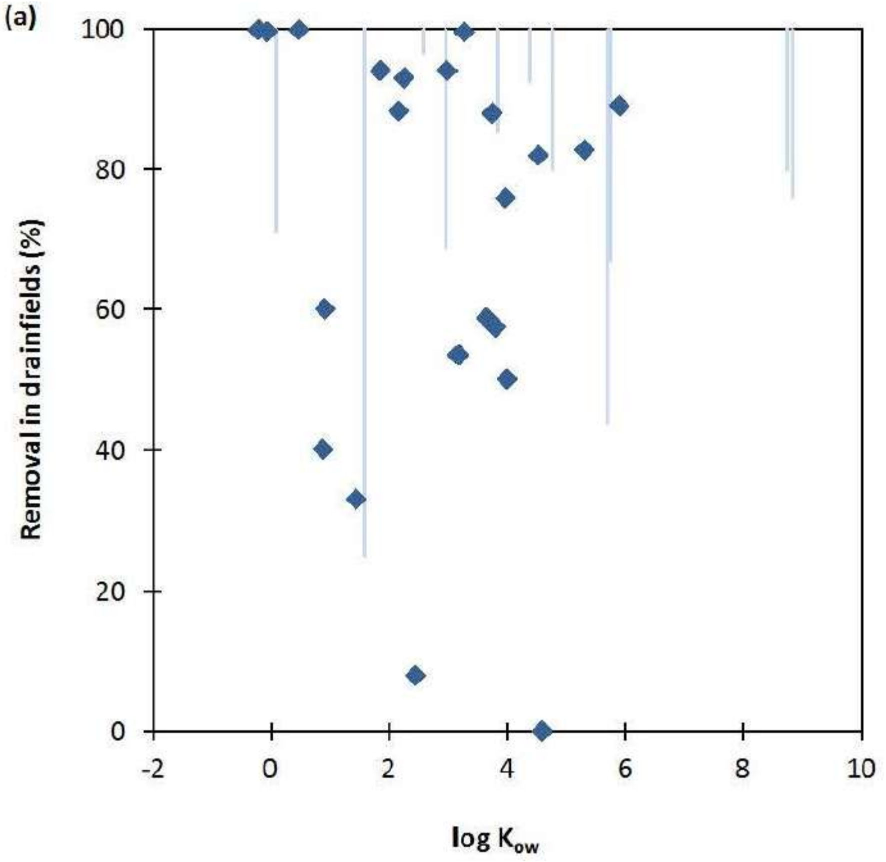
OWC median removal efficiencies in onsite drainfields and in WWTPs. For median removal efficiencies above a censoring value, the range of possible values is plotted as a light blue line.

Laurel A. Schaider; Kathryn M. Rodgers; Ruthann A. Rudel; *Environ. Sci. Technol.* 2017, 51, 7304-7317.

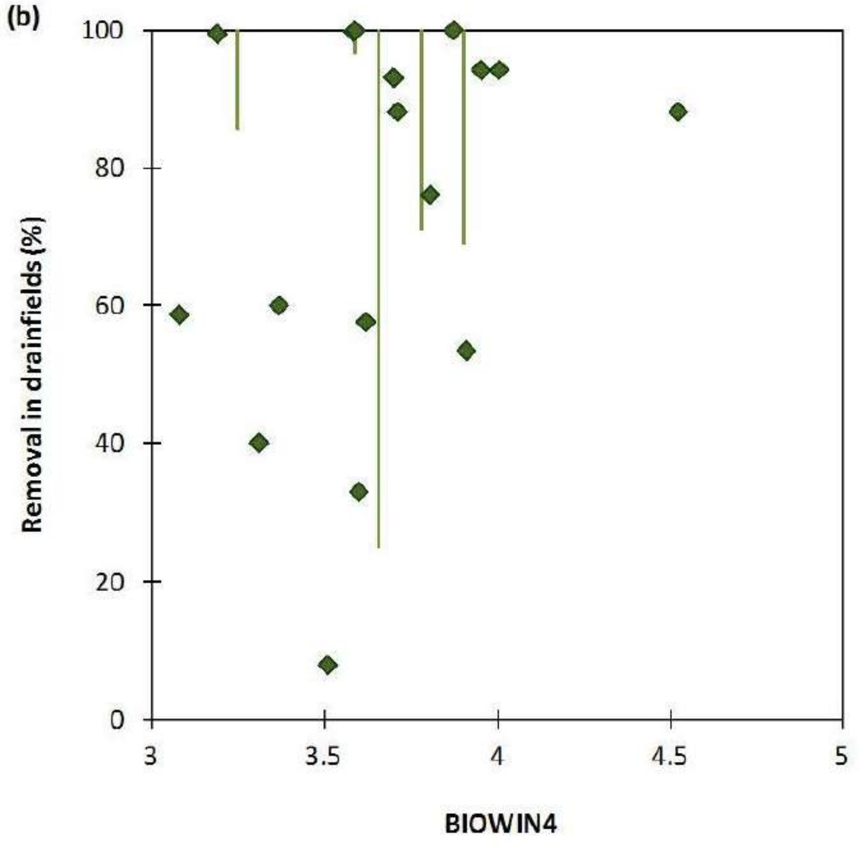
Laurel A. Schaider; Kathryn M. Rodgers; Ruthann A. Rudel; *Environ. Sci. Technol.* 2017, 51, 7304-7317.

Modeling approach applied to OWCs

Sorption



Biodegradation



<u>Fluorinated hydrocarbons</u>: nomenclature

- Poly- and Perfluoroalkyl substances (PFAS)
  - **<u>Per</u>** means all hydrogens are substituted with fluorine atoms
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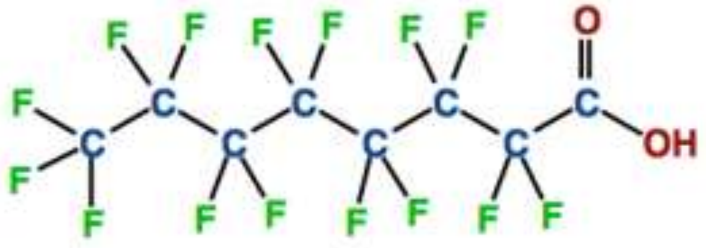
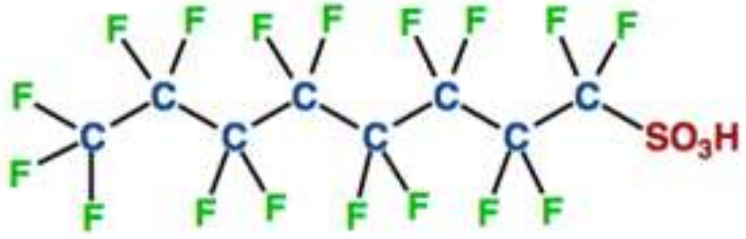


PFOA - perfluorooctanoic acid

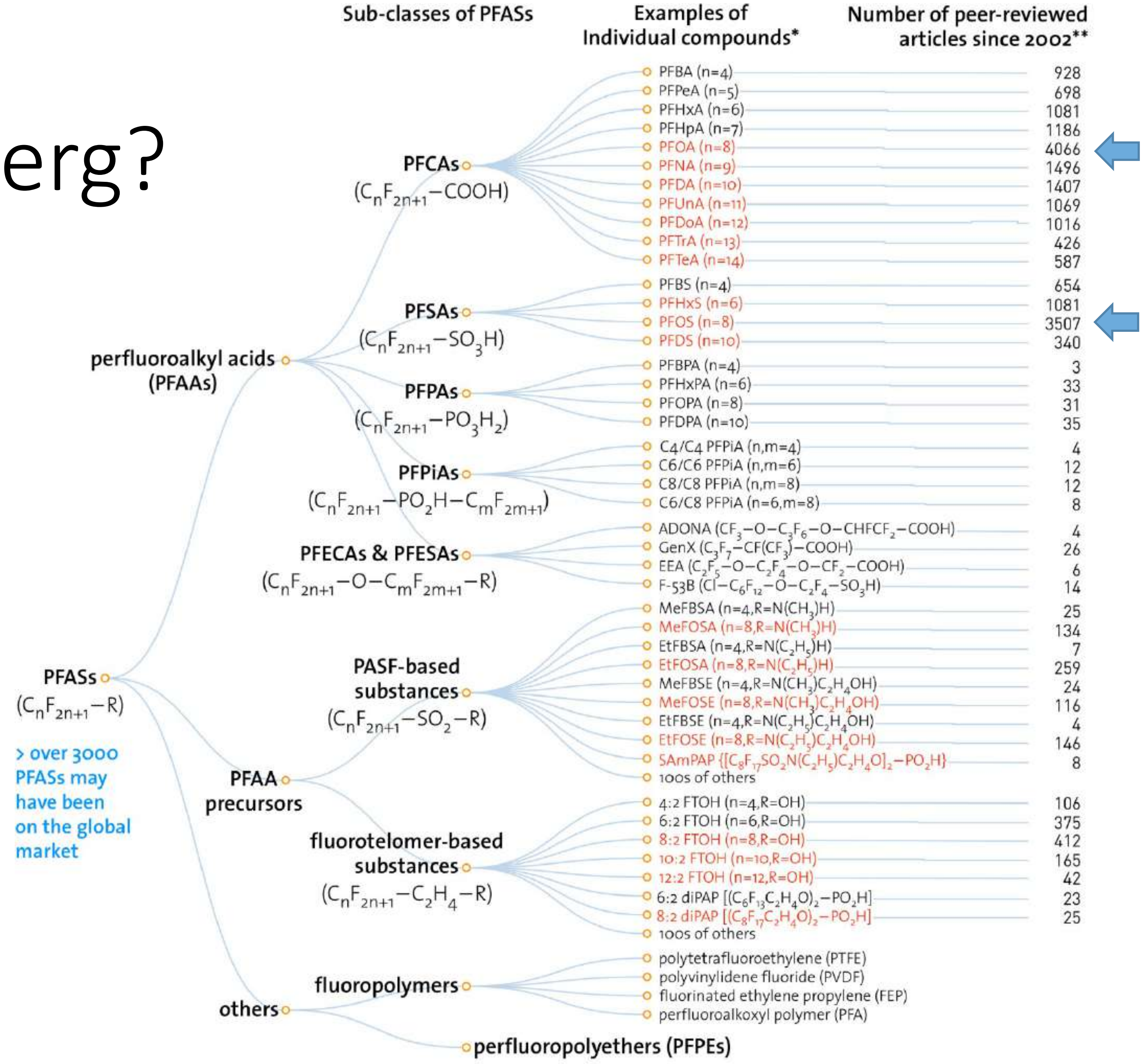


PFOS - perfluorooctanesulfonic acid

Tip of the PFAS-iceberg?

- Family tree of PFASs
  - most of the studies to date have focused on
    - Long-chain perfluoro carboxylic acids (PFCAs) which include PFOA
    - Long chain Perfluoro sulfonic acids (PFSAs), which include PFOS
    - Along with their major precursors.
  - New interest in GenX and others

Zhanyun Wang; Jamie C. DeWitt; Christopher P. Higgins; Ian T. Cousins; *Environ. Sci. Technol.* 2017, 51, 2508-2518



* PFASs in RED are those that have been restricted under national/regional/global regulatory or voluntary frameworks, with or without specific exemptions (for details, see OECD (2015), Risk reduction approaches for PFASs. http://oe.cd/1AN).

** The numbers of articles (related to all aspects of research) were retrieved from SciFinder® on Nov. 1, 2016.

PFAS properties

- High solubility, low volatility, low reactivity

$K_d = K_{oc} f_{oc}$

Compound	pKa	Vapor Pressure	Henry's Law Const	Aqueous Solubility	Log K_{oc} (L/Kg)	Degradation
PFOA $C_8HF_{15}O_2$	1.3	0.1 kPa (20°C) 10 mm Hg (25°C)	5.0 mol dm^{-3} atm^{-1} log H_e = -3.7 (-9.4 @pH7)	4.1 g/L (22°C) 9.5 g/L (25°C)	2.06 HL (Log K_dm = -1.54)	Stable
PFOS $C_8F_{17}SO_3^-$		3.31 x 10^4 Pa at 20°C		570 mg/L	2.57 HL (Log K_dm = -1.03)	Stable
PFHxS $C_6F_{13}SO_3$		0.61Pa (25°C) ES		6.2 mg/L ES 22 mg/L ES	3.5 ES (Log K_dm = -0.1)	Stable
PFBS $C_4F_9SO_3$		0.29 mm Hg at 20°C		8900 mg/L ES 344mg/L ES	2.2 ES 1.9 ES (Log K_dm = -1.5)	Stable
6:2 FTS $CF_3(CF_2)_5CH_2CH_2SO_3$		0.115Pa(25°C) ES 0.00086 mm Hg (25°C) ES		11 mg/L ES 2mg/L ES	4.0 ES (Log K_dm = -0.4)	Biodegradable

FTS=Fluorotelomersulfonic acid

Some data from: Michelle Crimi, Clarkson University

HL= from Higgins CP, Luthy RG. Sorption of perfluorinated surfactants on sediments. ES&T. 2006;40(23):7251-6.
ES = estimated from EPISuite (U.S. EPA http://www.epa.gov/opptintr/exposure/pubs/episuite.htm)

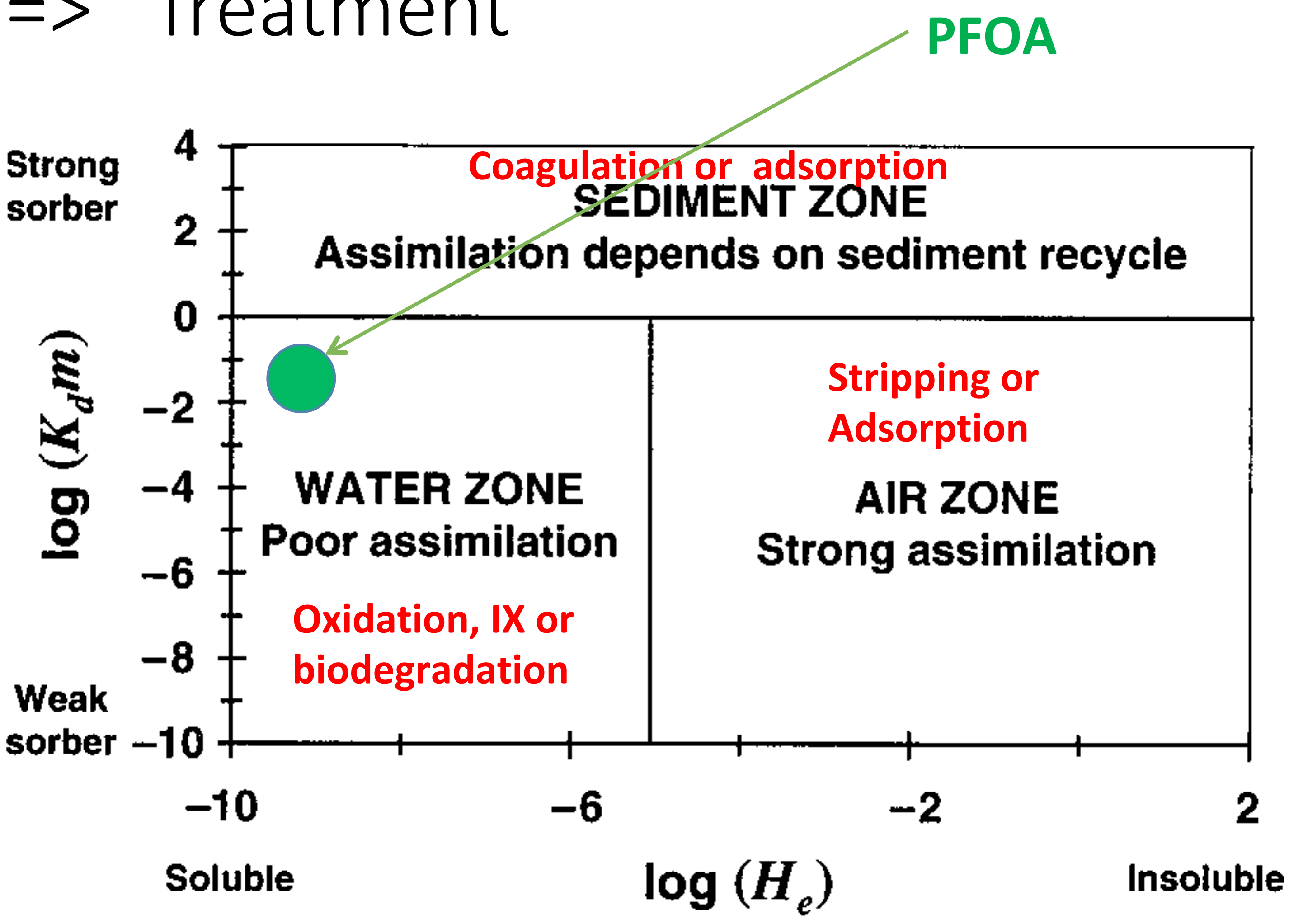
Chemical Properties => Treatment

- Fate in Aquatic systems
  - Relates to fate in treatment
- Partitioning Parameters

  $f_{oc} = 0.05$ $m = 5\ g/m^3$

  $K_d = K_{oc} f_{oc}$

- Aquatic assessment assuming:
  - T_a=283 K
  - M=200 g/mole
  - U_w = 5 mph
  - v_s =91 m/yr
- Assimilation refers to general rate of removal

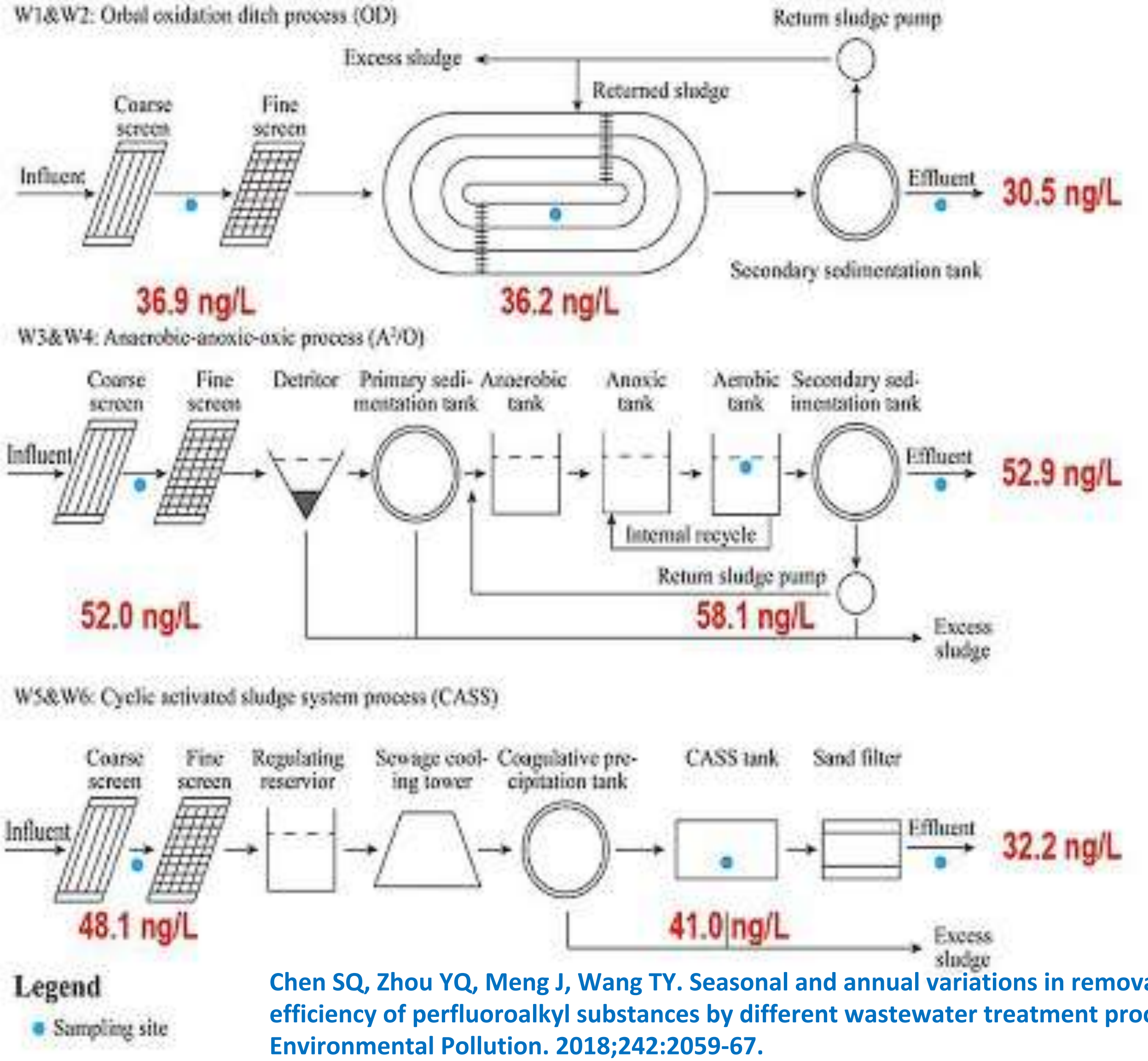


H_e units => atm m^3 $mole^{-1}$

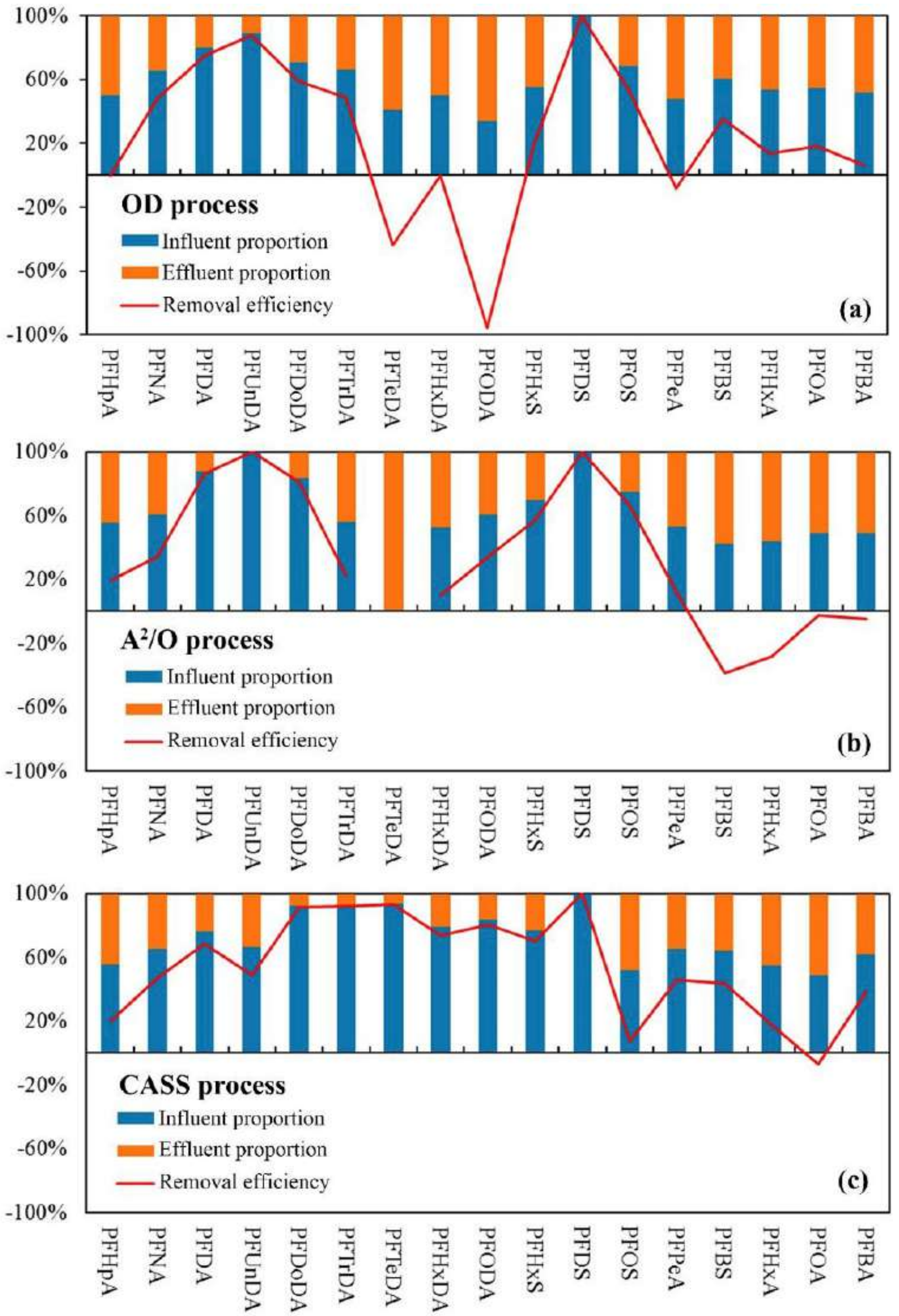
Chapra, 1991 [ASCE JEED 17(5)656]

Study of 6 WWTPs

Wastewater treatment processes

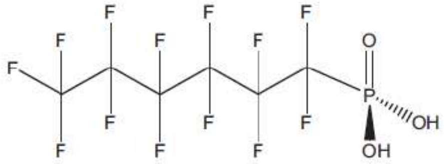
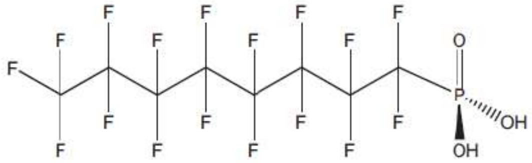
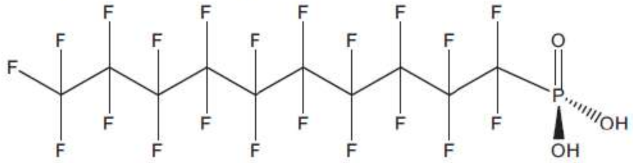
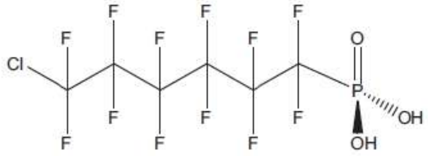


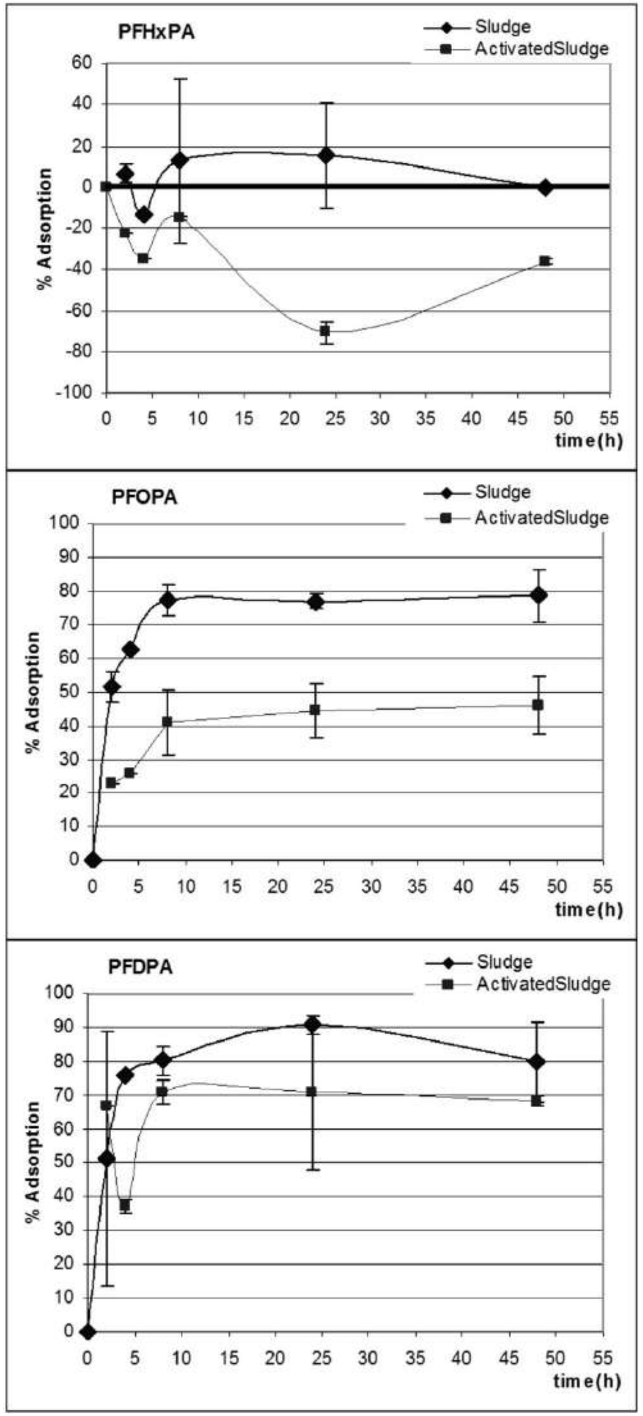
Chen SQ, Zhou YQ, Meng J, Wang TY. Seasonal and annual variations in removal efficiency of perfluoroalkyl substances by different wastewater treatment processes. Environmental Pollution. 2018;242:2059-67.



Sorption to biosolids in WWT

- On “activated sludge and final sludge”
- Aeration basin solids and secondary settled solids?

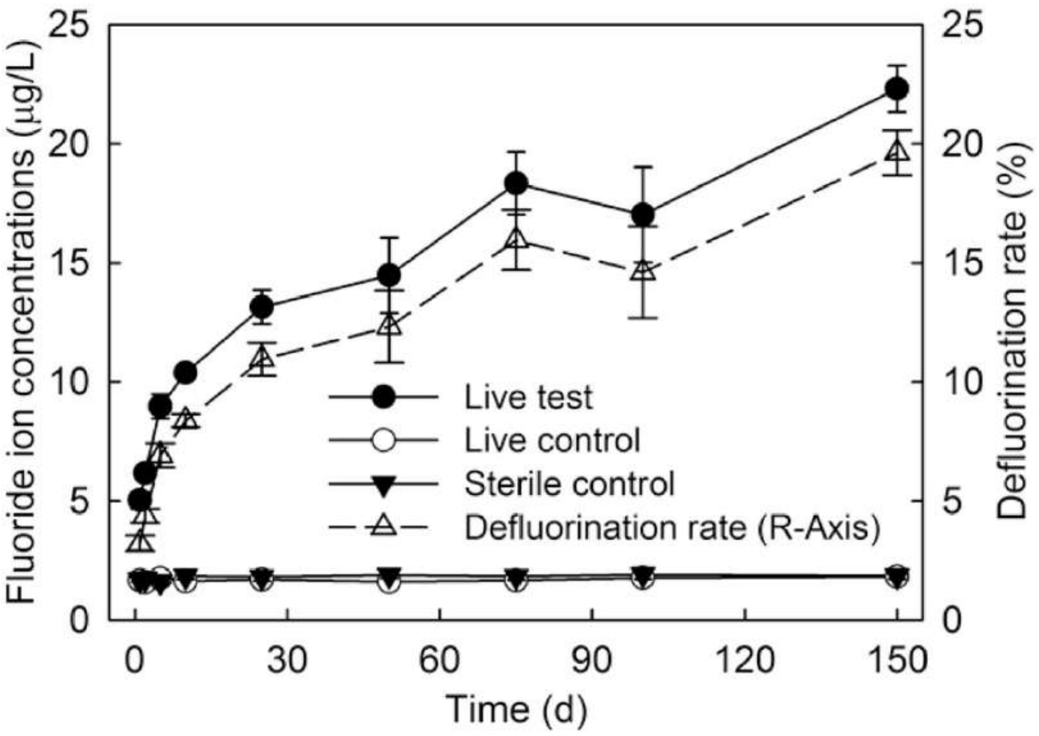
	MW (amu)	Structure
PFHxPA	400	
PFOPA	500	
PFDPA	600	
Cl-PFHxPA	416	



Llorca M, Farre M, Sanchez-Melsio A, Villagrasa M, Knepper TP, Barcelo D. Perfluoroalkyl phosphonic acids adsorption behaviour and removal by wastewater organisms. Science of the Total Environment. 2018;636:273-81.

Fluorotelamers in AS

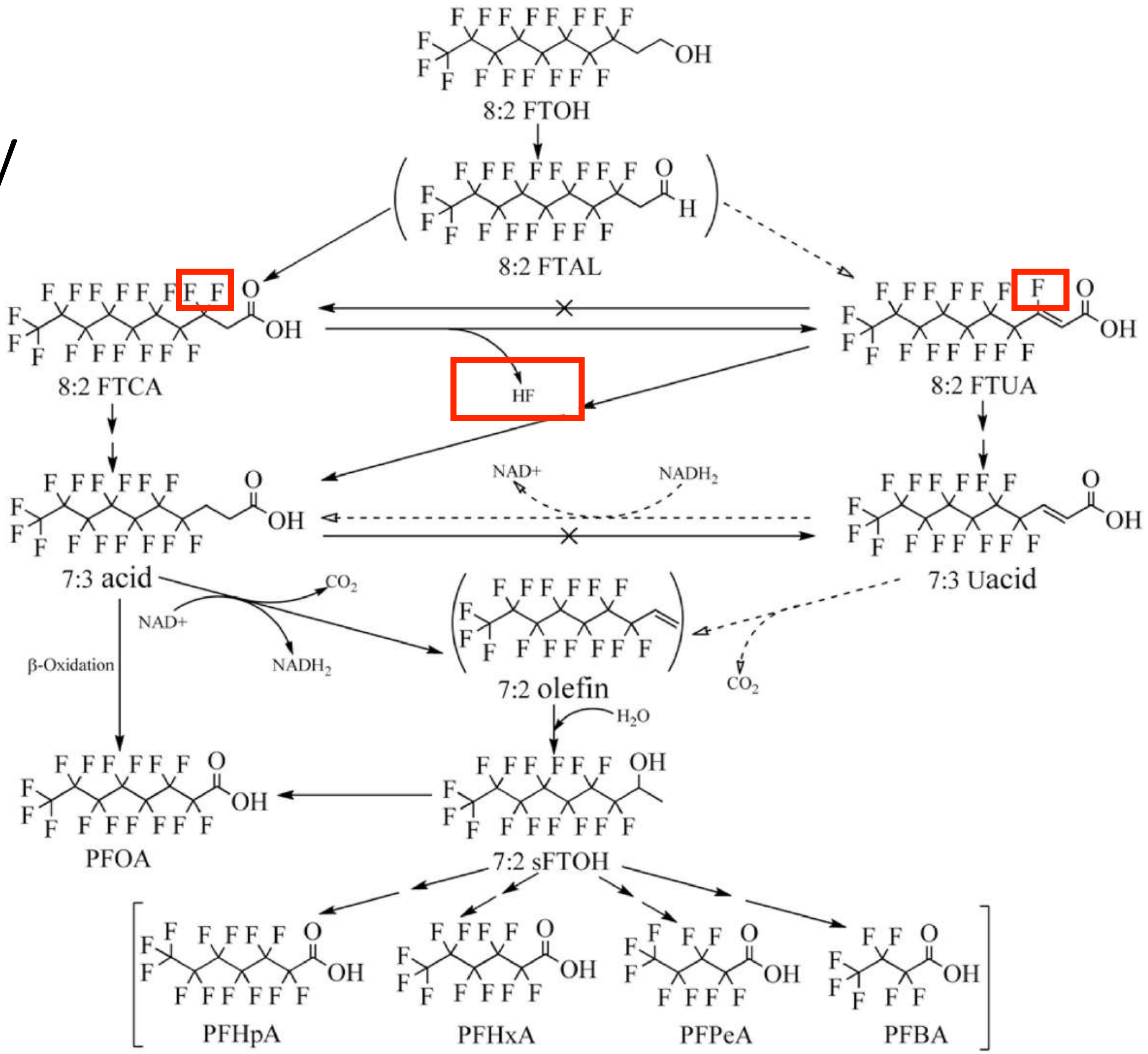
- Degradation under anaerobic conditions



Li F, Su QF, Zhou ZM, Liao XB, Zou J, Yuan BL, et al. Anaerobic biodegradation of 8:2 fluorotelomer alcohol in anaerobic activated sludge: Metabolic products and pathways. Chemosphere. 2018;200:124-32.

Anaerobic Pathway

- Formation of PFAS products
- Some de-fluorination!



Li F, Su QF, Zhou ZM, Liao XB, Zou J, Yuan BL, et al. Anaerobic biodegradation of 8:2 fluorotelomer alcohol in anaerobic activated sludge: Metabolic products and pathways. Chemosphere. 2018;200:124-32.

Impacts on WWT organisms

- Anecdotal evidence and growing literature

    - Chen HB, Zhou YF, Hu XY, Tian K, Zhang JF. Effects of chlortetracycline on biological nutrient removal from wastewater. Science of the Total Environment. 2019;647:268-74.
    - Du BB, Wang RF, Yang QX, Hu H, Li XN, Duan XW. Impact of tetracycline on the performance and abundance of functional bacteria of a lab-scale anaerobic-aerobic wastewater treatment system. Biochemical Engineering Journal. 2018;138:98-105.
    - Hu ZT, Sun PD, Han JY, Wang RY, Jiao L, Yang PF, et al. The acute effects of erythromycin and oxytetracycline on enhanced biological phosphorus removal system: shift in bacterial community structure. Environmental Science and Pollution Research. 2018;25(10):9342-50.
    - Liu H, Yang YK, Sun HF, Zhao L, Liu Y. Effect of tetracycline on microbial community structure associated with enhanced biological N&P removal in sequencing batch reactor. Bioresource Technology. 2018;256:414-20.
    - Wang K, Gao DD, Xu JR, Cai L, Cheng JR, Yu ZX, et al. Interaction of ciprofloxacin with the activated sludge of the sewage treatment plant. Environmental Science and Pollution Research. 2018;25(35):35064-73.
    - Yu NL, Zhao CK, Ma BR, Li SS, She ZL, Guo L, et al. Impact of ampicillin on the nitrogen removal, microbial community and enzymatic activity of activated sludge. Bioresource Technology. 2019;272:337-45.

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Only a problem at high Concentrations?

- Process performance
  - Decreasing rate of O_2 utilization
    - Ampicillin (20 mg/L)
  - Decreasing rate of nitrification
    - Ciprofloxacin (0.2 mg/L), Ampicillin (20 mg/L), tetracycline (5 mg/L)
  - Inhibition of denitrification
    - Chlorotetracycline
  - Poorer removal of orthophosphate
    - Chlorotetracycline (10 mg/L), erythromycin (5 mg/L)
- Shifting microbial ecology
  - Loss of accumulibacter, increase in competibacter
    - erythromycin (5 mg/L)
- Sludge behavior
  - Reduction in attached biomass and floc size
    - Ciprofloxacin (0.2 mg/L),
  - Sludge bulking
    - tetracycline (5 mg/L)
- Enzymatic impacts
  - Dehydrogenase inhibition
    - Ampicillin
  - Reductase
    - Chlorotetracycline
- Reactive Oxygen Species (ROS)
  - Increased production

Literature Cited

- Chen HB, Zhou YF, Hu XY, Tian K, Zhang JF. Effects of chlortetracycline on biological nutrient removal from wastewater. Science of the Total Environment. 2019;647:268-74.
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Boston Globe

March 10, 2008; page 2

released today, shows a growing urgency about climate change even within groups that once dismissed assertions of an overheating planet as a liberal ruse. The president of the Southern Baptist Convention was among the signers. (AP)

MICHIGAN

Paper ties mayor's friend to contracts

DETROIT — Companies headed by a friend of Mayor Kwame Kilpatrick won millions of dollars in city contracts while the friend secretly consulted with the mayor's chief of staff about bidding strategies, the Detroit Free Press reported yesterday. The paper said Bobby Ferguson and companies he partnered collected at least $45 million. Ferguson, the city, and a lawyer for former chief of staff Christine Beatty denied wrongdoing. (AP)

FLORIDA

Shuttle cleared for launch with new lab

CAPE CANAVERAL — NASA cleared the space shuttle Endeavour yesterday for launch early tomorrow to begin attaching a Japanese laboratory to the International Space Station and install Canadian-built robot arms. Liftoff from the Kennedy Space Center is scheduled for 2:28 a.m. Meteorologists predicted a 90 percent chance that weather conditions would be suitable for the rare night launch. (Reuters)

Bus, pickup collide, killing woman

FORT LAUDERDALE — A tour

Pharmaceuticals found in US drinking water

Trace quantities could endanger wildlife, humans

By Jeff Donn
ASSOCIATED PRESS

EPA ADDRESSING THE ISSUE

'We recognize it is a growing concern, and we're taking it very seriously,' said Benjamin Grumbles, assistant administrator for water at the federal EPA.

NEW YORK — An array of pharmaceuticals — including antibiotics, anticonvulsants, mood stabilizers, and sex hormones — have been found in the drinking water supplies of at least 41 million Americans, an Associated Press investigation found.

The concentrations of these pharmaceuticals are tiny, measured in quantities of parts per billion or trillion, far below the levels of a medical dose. And utilities insist that their water is safe.

But the presence of so many prescription drugs — and over-the-counter medicines like acetaminophen and ibuprofen — in so much of our drinking water is heightening worries among scientists of long-term consequences to human health.

In the course of a five-month inquiry, the AP discovered that drugs have been detected in the drinking water supplies of 24 major metropolitan areas — from southern California to northern New Jersey, from Detroit to Louisville, Ky.

Water providers rarely disclose results of pharmaceutical screenings, unless pressed, the Associated Press found.

For example, the head of a group representing major California suppliers said the public "doesn't know how to interpret the information" and might be unduly alarmed.

When people take pills, their bodies absorb some of the medication, but the rest of it passes through and is flushed down the toilet. The wastewater is treated before it is discharged into reservoirs, rivers, or lakes.

Then, some of the water is cleansed again at drinking water treatment plants and piped to consumers. But most treatments do not remove all drug residue.

While researchers do not yet understand the exact risks from decades of persistent exposure to random combinations of low levels of pharmaceuticals, recent studies, which have gone virtually unnoticed by the public, have found alarming effects on human cells and wildlife.

"We recognize it is a growing concern, and we're taking it very seriously," said Benjamin H. Grumbles, assistant administrator for water at the US Environmental Protection Agency.

The Associated Press reviewed hundreds of scientific reports, analyzed federal drinking water databases, visited environmental study sites, and treatment plants and interviewed more than 230 officials, academics, and scientists.

They also surveyed the nation's 50 largest cities and a dozen other major water providers, as well as smaller community water providers in all 50 states.

Here are some of the key test results:

■ Officials in Philadelphia said testing discovered 56 pharmaceuticals or byproducts in treated drinking water, including medicines for pain, infection, high cholesterol, asthma, epilepsy, mental illness, and heart problems. Sixty-three pharmaceuticals or byproducts were found in the city's watersheds.

■ Antiepileptic and antianxiety medications were detected in a portion of the treated drinking water for 18.5 million people in southern California.

■ Researchers at the US Geological Survey analyzed a Passaic Valley Water Commission drinking water treatment plant, which serves 850,000 people in northern New Jersey, and found a metabolized angina medicine and the mood-stabilizing carbamazepine in drinking water.

■ A sex hormone was detected in San Francisco's drinking water.

■ The drinking water for Washington, D.C., and surrounding areas tested positive for six pharmaceuticals.

The federal government doesn't require any testing and hasn't set safety limits for drugs in water. Some providers screen only for one or two pharmaceuticals, leaving open the possibility that others are present.

Of the 62 major water providers contacted, the drinking water for 28 was tested. Boston is among the 34 that haven't been tested, along with Baltimore, Chicago, Houston, Miami, New York, and Phoenix.

The investigation also indicates that watersheds, the natural sources of most of the nation's water supply, also are contaminated. Tests were conducted in the watersheds of 35 of the 62 major providers surveyed by the Associated Press and pharmaceuticals were detected in 28.

Yet officials in six of those 28 metropolitan areas said they did not go on to test their drinking water: Fairfax, Va.; Montgomery County in Maryland; Omaha; Oklahoma City; Santa Clara, Calif.; and New York City.

Of the 28 major metropolitan areas where tests were performed on drinking water supplies, only Albuquerque; Austin, Texas; and Virginia Beach, Va., said tests were negative.

Calif. dunes lure off-road enthusiasts, smugglers

By Richard Marosi
LOS ANGELES TIMES

of fortification that they hope will cut down on incursions.

the Department of Homeland Security seems willing to flex its