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Center for  
**Environmental Security**

**Sewage Metrology/Epidemiology  
by Example of the Antimicrobials  
Triclosan and Triclocarban**

**Rolf Halden, PhD, PE**  
Center for Environmental Security, Director  
The Biodesign Institute, Arizona State University

Department of Environmental Health Sciences, Adjunct Faculty  
Johns Hopkins University, Bloomberg School of Public Health

with **Arjun Venkatesan, PhD, EIT**

NACWA Webinar  
Broadcast on May 28, 2014

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**Outline**

- Contributors to this Presentation
- Rationale & Methodology
  - Archived Samples – ASU Human Health Observatory
  - Sewage Metrology
  - Mass Balances & Chemical Inventories
- Results
  - TCS & TCC
  - Other Contaminants of Emerging Concern (CECs)
- Questionnaire
- Summary & Conclusions
- Acknowledgements & References

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**Center for Environmental Security**

**This Presentation Contains Published Data from the Following Authors/Individuals from ASU**

Dr. Kristin McClellan  
 Dr. Jochen Heidler  
 Bipin Chari, M.S.  
 Evelyn Walters, M.S.



Dr. Arjun Venkatesan



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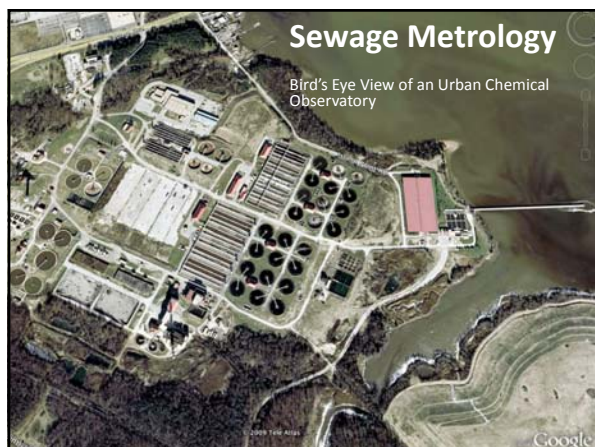
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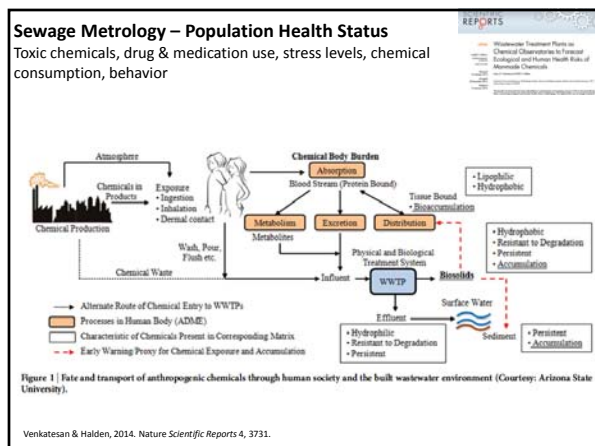
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## ASU Human Health Observatory (H2O)

### U.S. National Sewage Metrology Sample Repository

- >160 U.S. cities
- >10% of U.S. population
- Time points thus far: 2001, 2006/7, present
- >200 U.S. WWTPs total
- Representative of 16,000+ U.S. WWT plants



Venkatesan, Done and Halden, 2014 *Environmental Science and Pollution Research* (In Press)

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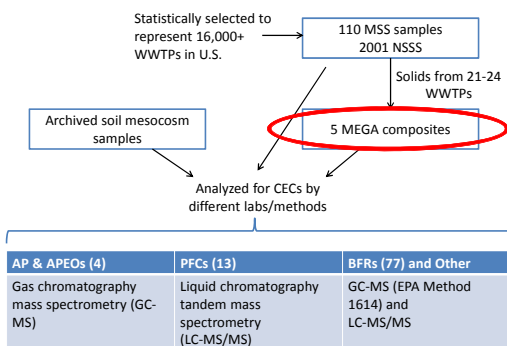
## H2O Repository at ASU:

### U.S. Geographic Coverage with WWTPs



Venkatesan, Done and Halden, 2014 *Environmental Science and Pollution Research* (In Press)

## Methodology



9

## Methodology – Studying the Persistence and Fate of CECs in Sludge-amended Outdoor Soil Mesocosms

- 2005 – 2008
- Baltimore, Maryland
- MSS/soil mixtures (1:2)
- Kept outdoors & sampled over 3 years



Venkatesan & Halden, 2014. Loss and in situ production of perfluoroalkyl chemicals in outdoor biosolids-soil mesocosms. In Press in *Environmental Research*

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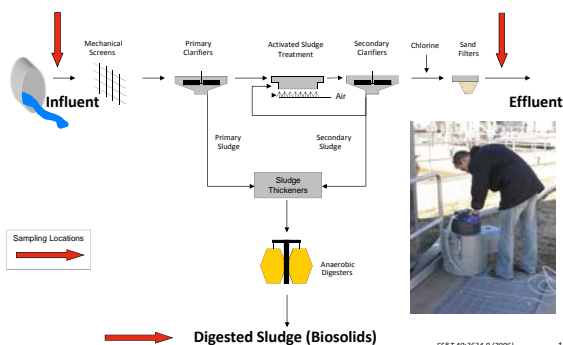
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## Sewage Treatment Plants

as Chemical Observatories by Example of Triclocarban & Triclosan



ES&amp;T 40:3634-9 (2006)

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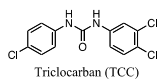
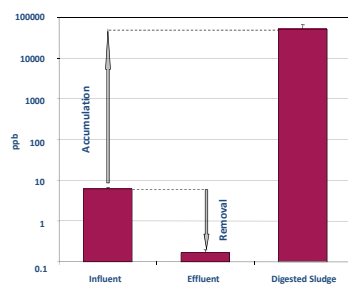
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## Removal ≠ Degradation



ES&amp;T 40:3634-9 (2006)

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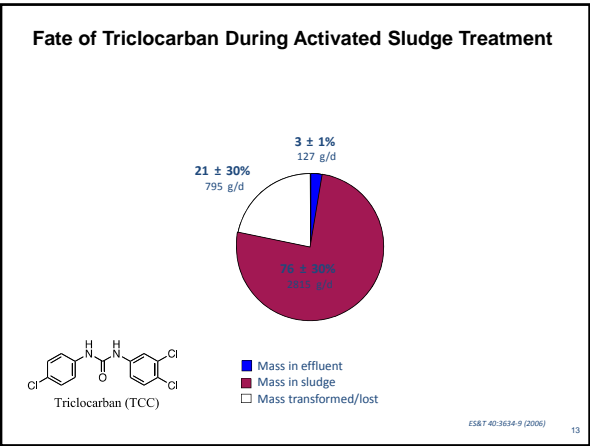
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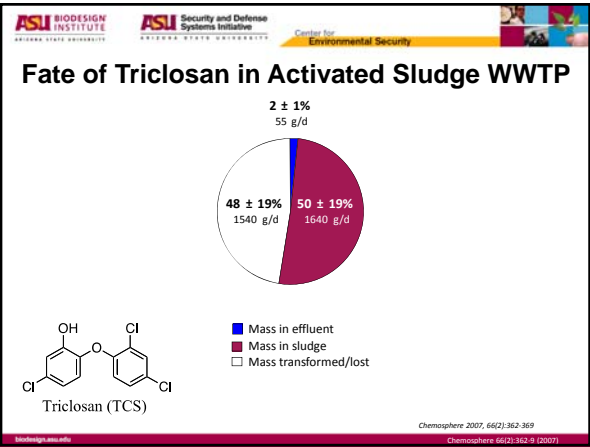
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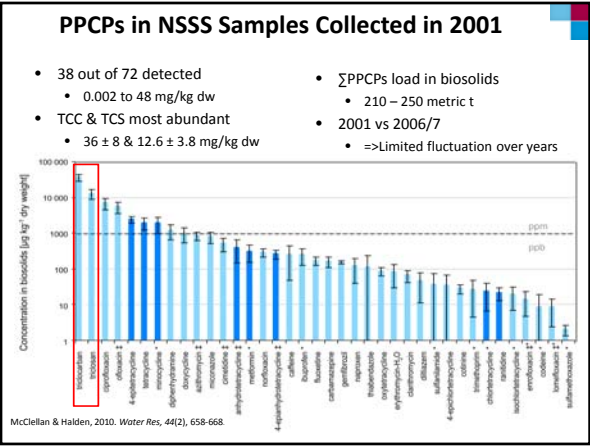
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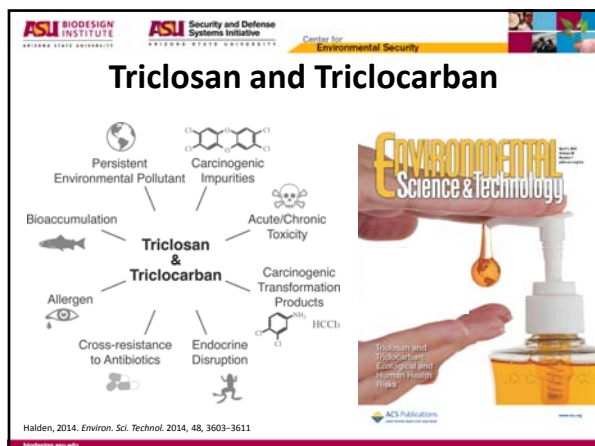
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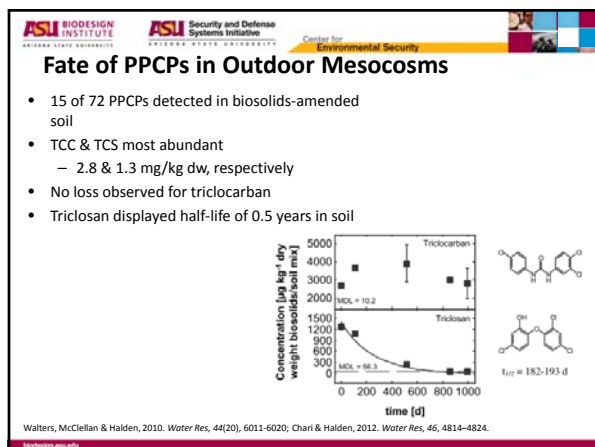
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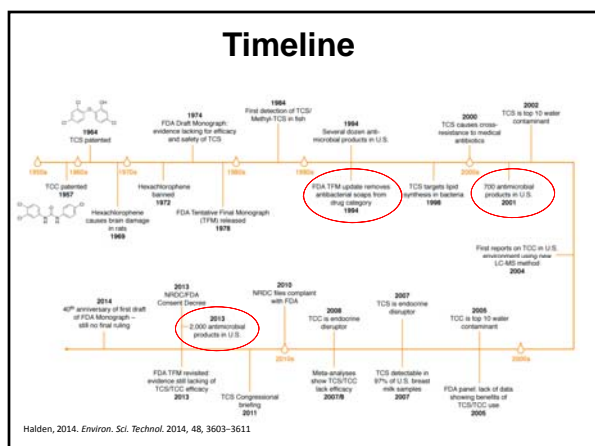
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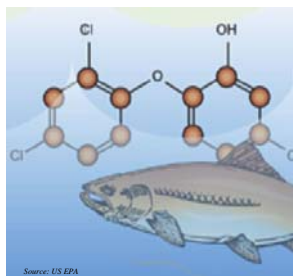
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## Contact Time is Key



~6 Seconds  
(ineffective)



Lifetime toxic  
exposure of aquatic  
organisms  
(toxic)

Halden, 2014. *Environ. Sci. Technol.* 2014, 48, 3603-3611

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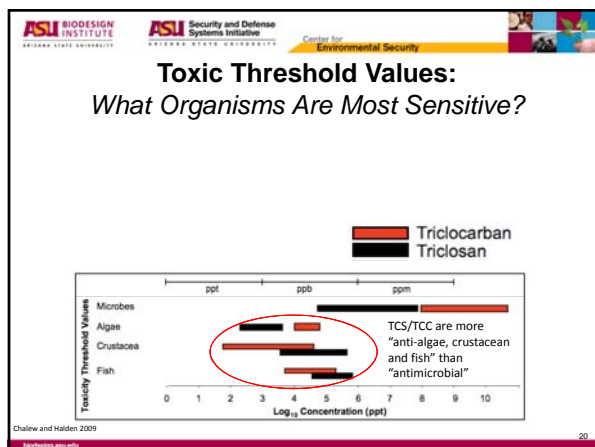
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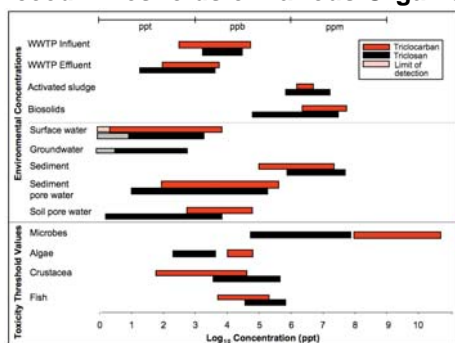
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## Concentrations Extant in U.S. Environment Exceed Thresholds of Various Organisms




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## Where Does the Sludge Go?

Used by consumers      Sequestered in sludge      Applied onto soils

ES&T 401(1) 3634-35, (2004)

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## Contaminated Land

Approximately 57 000 ± 233 000 kg/yr of Triclosan and 140 000 ± 211 000 kg/yr of Triclocarban are applied inadvertently on U.S. agricultural land as a result of sewage sludge disposal

Potential pathway for contamination of water and food with antimicrobials and drug-resistant microbes

Halden, 2014. *Environ. Sci. Technol.* 2014, 48, 3603-3611

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## Triclosan & Triclocarban: Key Sludge Pollutants

TCS and TCC are only two of 72 drugs monitored by EPA Method 1694

Yet, these two antimicrobials account for >60% of the **mass** of all drugs detectable in sewage sludge

McClellan & Halden, *Water Res.* 44: 626-636 (2010)

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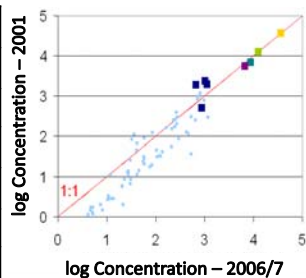
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Chemistry of Biosolids Collected in 2001 vs. 2006/7

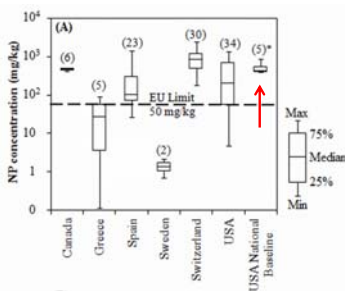
Compound	2007 Mean Concentration [ppm]	2001 EPA TNSSS Mean Concentration [ppm]
Triclocarban	36	39
Triclosan	12	16
Ciprofloxacin	7	10
Ofloxacin	5	8
Cimetidine, 4- Epitetracycline, Miconazole, Tetracycline	0.5 – 2.5	1 - 2



McClellan & Halden, Water Res. 44: 626-636 (2010) 25

First U.S. Nationwide Inventory for Alkylphenols & AP Ethoxylates in Sewage Sludge

- $656 \pm 272$  mg/kg dw
- 2,408 – 7149 t in sludge
- 1,204 – 4,289 t to U.S. soils
- $t_{1/2} = 301$ - 495 d
- U.S. levels exceed EU regulatory limits 10-fold
- U.S.: no regulatory limits

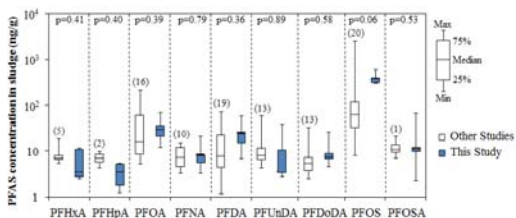


Venkatesan & Halden, 2013. Environ. Pollut. (174):189-193

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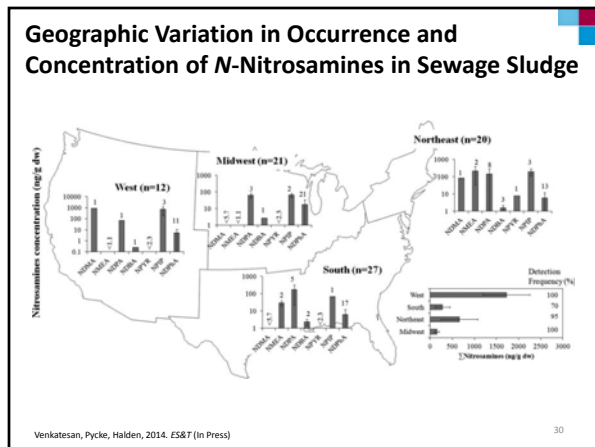
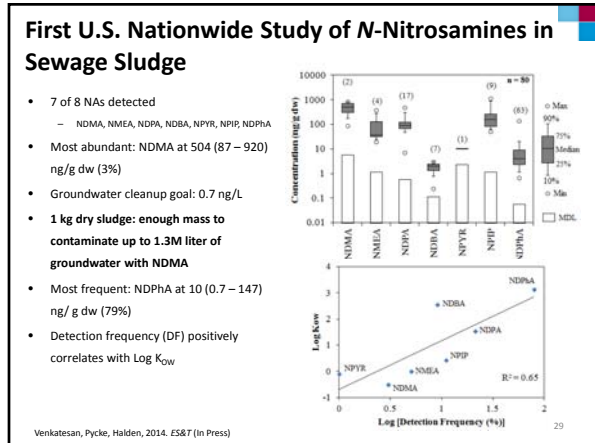
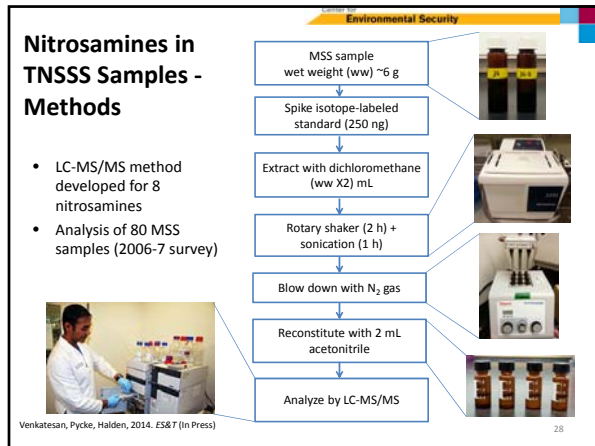
First U.S. Nationwide Inventory of Polyfluorinated Compounds in Sewage Sludge

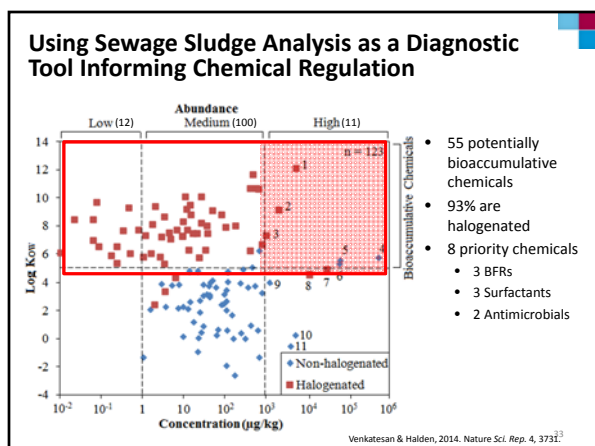
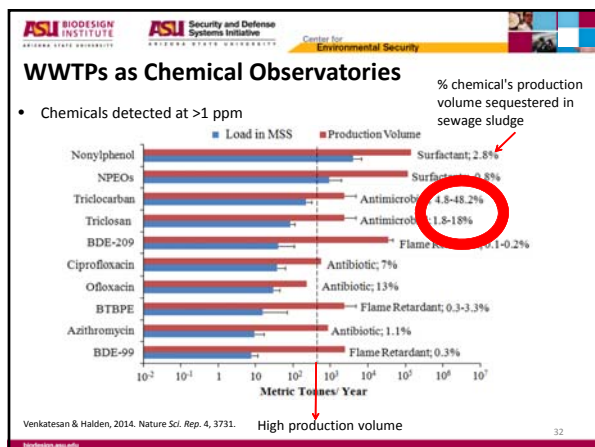
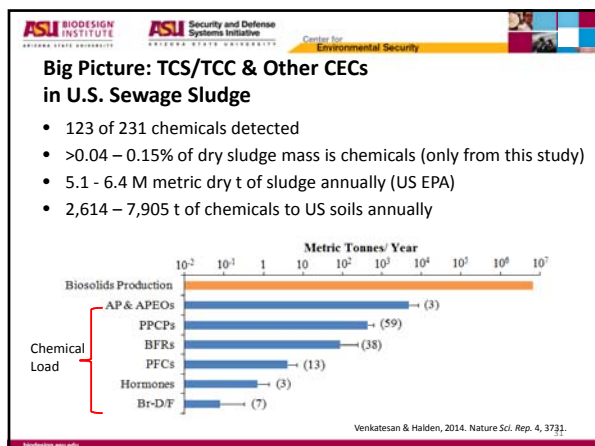
- 13 PFCs; 10 PFCs 100% detection
- PFOS:  $403 \pm 127$   $\mu$ g/kg dw; PFOA:  $34 \pm 22$   $\mu$ g/kg dw
- $\Sigma$ PFCs estimated at 2,749 - 3,450 kg/year
- No difference: pre-phase out (2001) and post phase-out (2006/7)

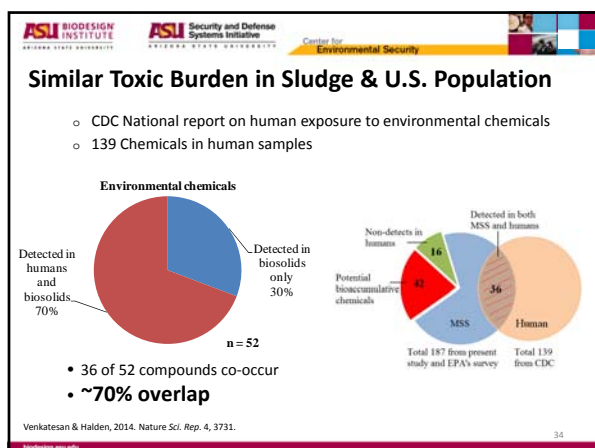


Venkatesan & Halden, 2013. J. Hazard. Mater. (252):413-418

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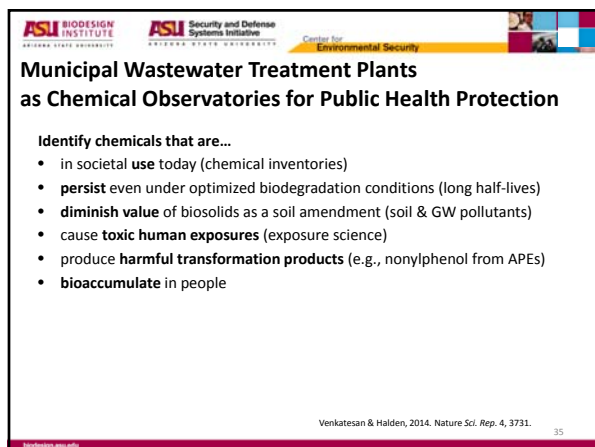
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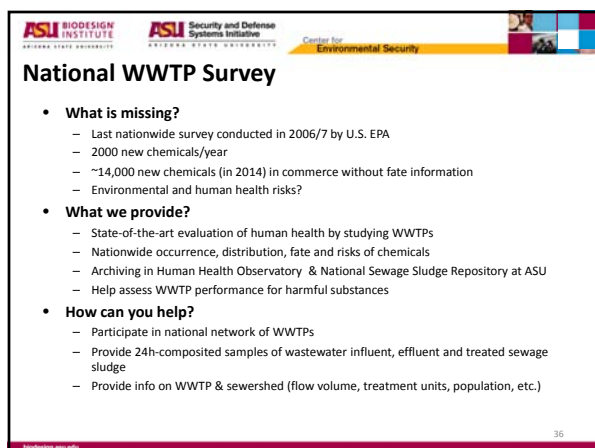
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### Contaminants Studied Thus Far

- Emerging contaminants
  - Pharmaceuticals and personal care products
  - Brominated flame retardants
  - Perfluorinated compounds
  - Hormones
  - Alkylphenol surfactants
  - Brominated dioxins and furans
  - Nitrosamines
- Future
  - More traditional and emerging contaminants
  - Human health indicators (biomarkers)
  - Illicit drugs
  - Biological agents

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### Questionnaire

- Do you want to know the performance of your WWTP?
- Would you like to participate in a nationwide WWTP survey?
- Please check any criteria that would prevent your participation in this survey:

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### Questionnaire (continued)

- Will you provide samples (influent/effluent/biosolids), under the condition that the identity of your facility is **NOT revealed**?
- Which sampling frequency would you participate in?

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## Summary & Conclusions

- TCS and TCC are examples of poorly designed, non-green chemicals featuring PBT characteristics (PBT= persistent, bioaccumulative, toxic)
- Sewage metrology can serve to inexpensively determine inventories of PB(T) chemicals and releases to soils nationwide
- Halogenated compounds pose particular concern
  - All Stockholm Convention compounds are organohalogens
  - 74 percent of SDWA regulated organic compounds carry at least one or more halogens
- Biosolids can harbor water-soluble pollutants that may leach into soil & groundwater
  - e.g., PPCs, carcinogenic nitrosamines including NDMA
- Monitoring chemicals in biosolids can identify problematic compounds and inform biomonitoring & regulatory actions
- ASU offers opportunity for you to learn more about your facility by participating in a nationwide WWTP survey

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## Acknowledgments

US EPA  
NIEHS  
CDC  
Rick Stevens  
Harry McCarty  
Randhir Deo  
Evelyn Walters  
Kristin McClellan  
Jochen Heidler  
AND MANY OTHERS

**NIH** National Institute of Environmental Health Sciences

**CDC**

**UNITED STATES - ENVIRONMENTAL PROTECTION AGENCY**

NIEHS 1R01ES015445  
NIEHS 1R01ES020889  
Johns Hopkins Center for a Livable Future

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- Venkatesan, A. B. F. G. Pyke, and R. U. Halden. 2014. Detection and Occurrence of N-Nitrosamines in Archived Biosolids from the Targeted National Sewage Sludge Survey of the U.S. Environmental Protection Agency. *Environmental Science & Technology* (In Press). dx.doi.org/10.1021/es5001352.
- Halden, R. U. 2014. On the Need and Speed of Regulating Triclosan and Triclocarban in the United States. *Environmental Science & Technology* 48:3603–3611. DOI: 10.1021/es500495p. *Environmental Science & Technology* Cover & Feature, April 1, 2014.
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- Venkatesan A. and R. U. Halden. 2014. Brominated flame retardants in U.S. biosolids from the EPA national sewage sludge survey and chemical persistence in outdoor soil mesocosms. *Water Research* 55, 133-142. <http://dx.doi.org/10.1016/j.watres.2014.07.021>
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Interested in the Nationwide WWTP Survey? Contact: halden@asu.edu

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### Poll

- Did you wash your hands with antibacterial soap today?
- Do you brush your teeth with Colgate Total?

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## Widespread Exposure

at least **1 million lbs** produced annually in US

**75%** Americans surveyed with triclosan in urine

**Dramatic rise** in blood levels after brushing with triclosan toothpaste for 14 days



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## Human Health Concerns



- Hormone disruption
- Weakened muscles
- Allergies and Asthma

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## Bacterial Impacts



### Antibiotic Resistance

- Potential to promote drug-resistant bacteria

### Beneficial Communities

- Microbiome
- Wastewater treatment plants

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## Unnecessary Exposure



No more effective than plain soap and water.

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"At this time, FDA does not have evidence that triclosan added to antibacterial soaps and body washes provides extra health benefits over soap and water."

**April 2010**

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## Initiatives

**Bans**

- Minnesota (2014)
- UT Austin (2012)
- Chicago (pending)

**Procurement**

- Minnesota (2013)
- Jamtland, Sweden (2010)
- San Jose, CA (2008)
- East Bay Municipal Utility District (2007)
- Palo Alto, CA (2006)

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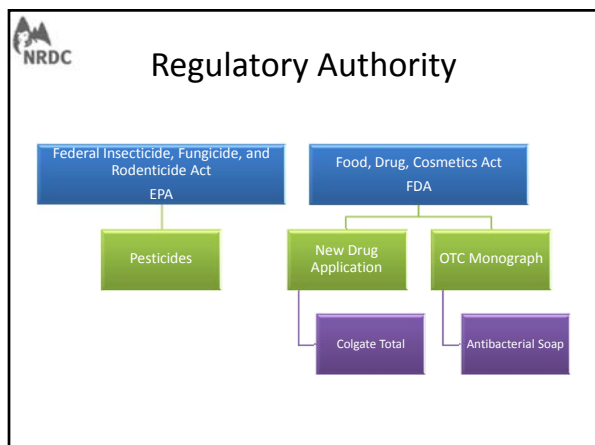
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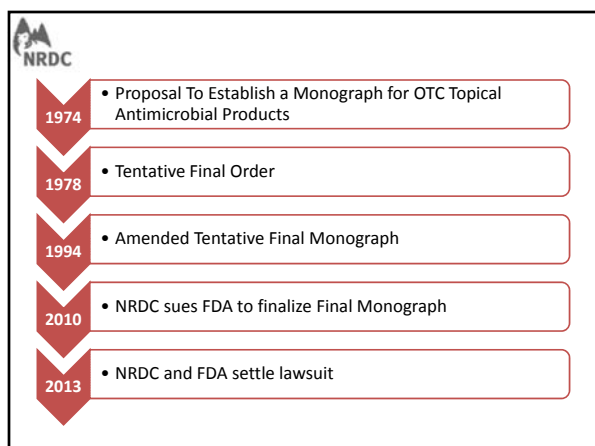
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2014

- June 16, 2014 – comments due on Tentative Final Monograph
- <http://www.regulations.gov/#!submitComment;D=FDA-1975-N-0012-0317>

2015

- February 17, 2015 – rebuttal comments due

2016

- September 15, 2016 – Final Monograph

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Natural Resources Defense Council  
[www.nrdc.org](http://www.nrdc.org)

NRDC Factsheet  
[www.nrdc.org/health/files/antimicrobials.pdf](http://www.nrdc.org/health/files/antimicrobials.pdf)

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**Thank You**

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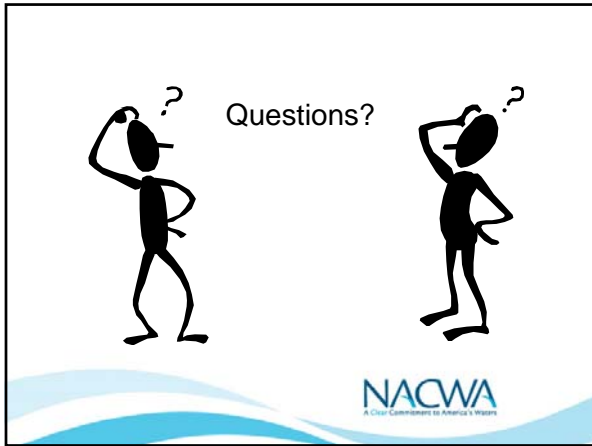
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