Pharmaceuticals in the Environment: 
*What we know and need to know*

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Chemicals of Emerging Environmental Concern: *Emerging Contaminants*

Not Just Pharmaceuticals!

Chemicals that:

- *interact* in their affect on organisms, &
- *co-occur* in the environment.
Modes of Action with Chemical Interactions

• Endocrine Disruption (biogenic hormones, synthetic hormones, hormone mimics & blockers)

• Antimicrobial Resistance (synthetic antimicrobials, natural antimicrobials, metals, pesticides?)
Emerging Contaminants

- Human & Veterinary Pharmaceuticals
- Detergents
- Antioxidants
- Fire retardants
- Disinfectants
- Fumigants
- Fragrances
- Pesticides/Repellants
- Industrial Chem’s, HPVs
- Some Metals
- Biogenic Hormones
- Phytoestrogens
- Natural antimicrobials
- Natural pesticides
- Degradates/Metabolites
Framing Research Questions

- Are Pharm’s entering our environment?
- What are the source pathways, their chemical signatures/loads?
- Are there sensitive environmental settings?
- Do Pharm’s have adverse ecological health effects?
- Do Pharm’s persist to finished drinking water and are they a human health risk?
- How can we minimize their entry to the environment or remove them?
Are Emerging Contaminants entering our natural environment?
National Surveys of “Susceptible” Waters

- **High density** of population and animal production.
- Present in water at **sub-ppb concentrations**
- Present in complex **mixtures**.
- Greater levels in streams than wells

*Kolpin, et al., 2002; Barnes et al., 2008*
Don’t just look in stream waters!

Furlong, et al., 2003
Measurement Capabilities

- Clofibric Acid (Buser 1998)
- 32 drugs in German WWTPs (Ternes 1998)
- 45 drugs in US Rivers (Kolpin et al. 2002)
- amphetamines
- antibiotics
- antidepressants (SSRIs)
- antiphlogistics
- antivirals
- barbiturates
- beta-blockers
- Ca channel blocker
- contraceptives
- cytostatics
- fibrates
- fibrates
- glucocorticoids
- muscle relaxants
- opioids
What are the source pathways to the environment and their chemical signatures/loads?
Human Source Pathways?

- WW Treatment Plants
- Domestic Septic Systems
- Land Application
- Industrial/Commercial Discharges
- Landfills
- Water Reuse
Animal Source Pathways?

- Grazing
- AFOs/CAFOs
- Waste lagoons
- Land application
- Processing
- Aquaculture
- Pets
WWTP Discharges to Streams

- Complex chemical mixtures.
- Multiple upstream WWTP discharges.
- Significant % WW in some streams (arid or urbanized watersheds).
ECs in Biosolids Destined for Land App

<table>
<thead>
<tr>
<th></th>
<th>Range</th>
<th>Median</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbemazepine</td>
<td>15-1,200</td>
<td>68</td>
</tr>
<tr>
<td>Diphenhydramine</td>
<td>32-22,000</td>
<td>340</td>
</tr>
<tr>
<td>Fluoxetine</td>
<td>100-4,700</td>
<td>370</td>
</tr>
<tr>
<td>Triclosan</td>
<td>1,170-32,900</td>
<td>10,200</td>
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</tbody>
</table>

- Biosolids from 9 sites
- 53 (of 87) ECs detected
- 30 to 45 in each biosolid
- 25 in all samples

Kinney et al., 2006
Septic Systems

ECs found in septic wastewaters and adjacent groundwater (Swartz et al., 2006; Carrara et al., 2008)

Higher levels in commercial than domestic septic systems (restaurants, convenience stores, retail centers, schools, veterinary hospitals) (Conn et al., 2006)
Conc. Animal Feeding Operations

• Large facilities
• Prophylactic doses
• Antibiotics, growth promoters, estrus modulators, ...
## Antibiotics in Swine Lagoons

### Antibiotics in 46 Swine AFOs

<table>
<thead>
<tr>
<th>Antibiotic</th>
<th>Detections (%)</th>
</tr>
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<tbody>
<tr>
<td>Chlorotet-total</td>
<td></td>
</tr>
<tr>
<td>Sulfamethazine</td>
<td></td>
</tr>
<tr>
<td>Lincomycin</td>
<td></td>
</tr>
<tr>
<td>Oxytetracycline</td>
<td></td>
</tr>
<tr>
<td>Sulfathiazole</td>
<td></td>
</tr>
<tr>
<td>Tetracycline</td>
<td></td>
</tr>
<tr>
<td>Tylosin</td>
<td></td>
</tr>
<tr>
<td>Sulfadimethoxine</td>
<td></td>
</tr>
<tr>
<td>Erythromycin-H2O</td>
<td></td>
</tr>
<tr>
<td>Trimethoprim</td>
<td></td>
</tr>
<tr>
<td>Virginiamycin</td>
<td></td>
</tr>
<tr>
<td>Sulfamethoxazole</td>
<td></td>
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<tr>
<td>Sulfame-thizole</td>
<td></td>
</tr>
</tbody>
</table>

Meyer et al., 2003
Are there sensitive environmental settings?
Hospital Effluents (Antibiotics)

3 Hospitals in Southeast Queensland, Australia

16 of the 27 antibiotics detected.

Watkinson et al., 2009
Pharm. Manufacturing Facilities

WWTP near Hyderabad India

• Receives effluent from 90 bulk drug manufacturers.
• Samples on consecutive days.

<table>
<thead>
<tr>
<th>Active Ingredient</th>
<th>Drug Type</th>
<th>Range (μg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ciprofloxacin</td>
<td>antibiotic</td>
<td>28,000-31,000</td>
</tr>
<tr>
<td>Losartan</td>
<td>angiotensin II receptor antagonist</td>
<td>2,400-2,500</td>
</tr>
<tr>
<td>Cetirizine</td>
<td>H₁-receptor antagonist</td>
<td>1,300-1,400</td>
</tr>
<tr>
<td>Metoprolol</td>
<td>B₁-andrenoreceptor antagonist</td>
<td>800-950</td>
</tr>
<tr>
<td>Enrofloxacin</td>
<td>Antibiotic</td>
<td>780-900</td>
</tr>
<tr>
<td>Citalopram</td>
<td>SSRI</td>
<td>770-840</td>
</tr>
<tr>
<td>Norfloxacin</td>
<td>antibiotic</td>
<td>390-420</td>
</tr>
<tr>
<td>Lomefloxacin</td>
<td>antibiotic</td>
<td>150-300</td>
</tr>
<tr>
<td>Enoxacin</td>
<td>antibiotic</td>
<td>150-300</td>
</tr>
<tr>
<td>Ofloxacin</td>
<td>antibiotic</td>
<td>150-160</td>
</tr>
</tbody>
</table>
Hydrologic Events

Spring Flush

Midwest Floods, Spring ‘08

21 herbicides, 27 degradates, 36 antibiotics in 51 Midwest Streams

Scribner et al., 2003
Animal Waste Lagoon Failure

Midwest Headwaters Stream
Do EC’s have adverse ecological health effects?
EC Uptake in Organisms

Plant Tissue

- Oxytetracycline in alfalfa (Kong et al., 2007).
- Oxytetracycline, flumequine & oxolinic acid in bryophytes (Delepee et al., 2004).
- Trimethoprim in carrots & lettuce (Boxall et al., 2006).
- Sulfamethazine in corn, lettuce, potatoes (Dolliver, et al., 2007).

Animal Tissue

- Fluoxetine in bluegill, catfish, carp, crappie (Brooks et al., 2005).
- Gemfibrozil in goldfish (Mimeault et al, 2005).
- Triclosan & Methyl-triclosan in carp (Leiker et al., 2008).
- Trimethoprim & Triclosan in earthworms (Kinney et al., 2008).
Ecological Effects

- **Antibiotics**: Reduced soil microbial activity at env. concentrations (Costanzo et al., 2005; Thiele-Bruhn and Beck, 2005).

- **Diclofenac (NSAID)**: Consumption of diclofenac-treated meat caused renal failure in *vultures* (Oaks et al., 2004).

- **Ciprofloxacin, triclosan, Tergitol NP 10**: shifts in algal community structure (Wilson et al., 2003).

- **Drug Mixtures 13**: Inhibited growth of human embryonic cells at environmental levels (Pomati et al., 2006, 2008).

- **Fluoxetine**: Affected reproduction in freshwater molluscs -- water-sediment exchange (Sanchez-Arguello et al., 2009).

- **Antidepressants**: (environmental levels) Affected predator avoidance behavior of larval fathead minnow (McGee et al. 2009).

- **4-nonylphenol**: (environmental exposures) Impaired reproductive potential of male fathead minnows. (Schoenfuss et al., 2008).

- **Alkylphenolethoxylates**: (environmental mixtures) Reduced reproductive competence in male fathead minnows (Bistodeau et al., 2006).

- **Sewage Sludge**: Affected bone homeostasis in sheep (Lind et al., 2009).
Endocrine Disruption: A Case Study

17b-Estradiol in stream water, in ng/L

Boulder Creek, CO

Boulder WWTP

Low-head Dam

Boulder Cr.

Dry Cr.

<0.8

2.9

2.1

1.2

1.4

Vajda et al., 2008
Estrogenicity of Boulder Effluent & Boulder Creek

Vajda et al., 2008

Spring 2005

Total Estrogen Equivalency (in ng L⁻¹ 17β-Estradiol)
Endocrine Effects in Boulder Ck (White Sucker)

Vajda et al., 2008

Intersex

Blood Vitellogenin

Cellular Abnormalities

Vajda et al., 2008
Do EC’s persist to finished drinking water and are they a human health risk?
Natural Dilution and Degradation

National Source Water Survey

- DW Sources (Streams) 49
- Susceptible Streams 139

Focazio et al., 2008
ECs in Finished Drinking Water

*11 of 20 Pharm’s not detected

Conducted by Southern Nevada Water Authority

Supported by AWWA Research Fnd. & WateReuse Fnd.

Benotti et al., 2008
Human Health Risk?

• Chronic low-level exposure.
• Exposure to chemical mixtures.
• Sensitive subpopulations.
• Can we prioritize chemicals systematically for effects studies?
How can we minimize their entry to the environment or remove them?
WWTPs Ability To Reduce ECs

Phillips et al., 2008
Removal in Treatment, NJ Facility

Levels Generally Reduced by Treatment with GAC Filters

Stackelberg et al., 2004 & 2007
Thank you!

And thanks to the many researchers who provided the information presented.

For more info on USGS EC research:
http://toxics.usgs.gov