



Chemicals (Contaminants) of Emerging Concern from Rural Nonpoint Sources

Kang Xia

Dept. Crop and Soil Environmental Sciences
Virginia Tech



VIRGINIA TECH.

What are CECs?

Human uses

- Prescription drugs
- over-the counter drugs
- Therapeutic drugs
- Veterinary drugs
- Fragrances
- Cosmetics
- Sun-screen products
- Diagnostic agents
- Nutraceuticals (e.g., vitamins)
- Illegal drugs
- Flame retardants
- Additives in consumer products



Animal production uses

- Therapeutic (disease control)
- Sub-therapeutic (growth promotion)



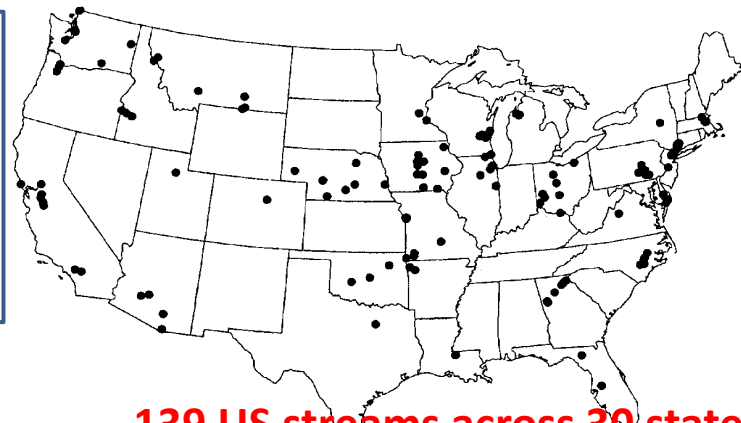
Widespread occurrence of CECs in aquatic systems

- **United States**

82 of the 95 ECs were found
ECs in 80% of the streams
ppt to several hundreds ppb
levels

(Environ. Sci. Technol, 2002, 36:1202-1211)

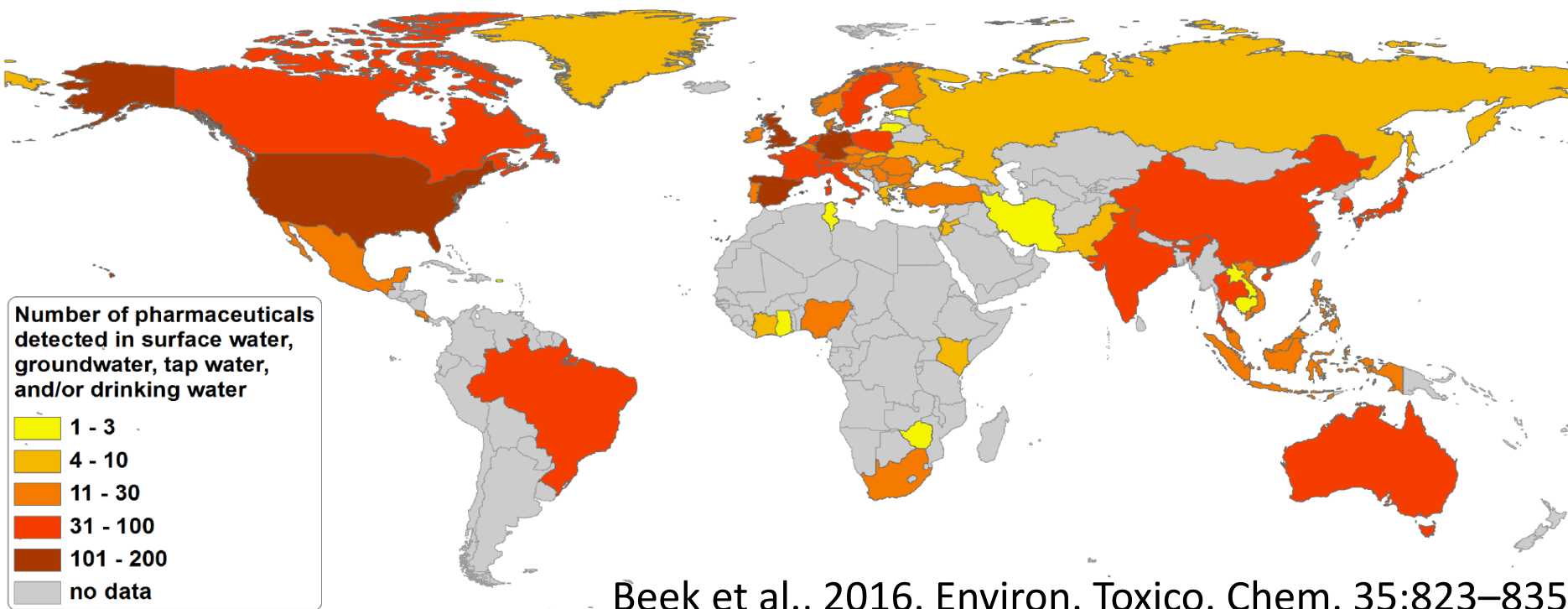
31 antibiotics
15 prescription drugs
7 nonprescription drugs
18 steroids and hormones
24 personal care-related
compounds



139 US streams across 30 states

- **Worldwide**

631 CECs have been detected in 71 countries



Beek et al., 2016. Environ. Toxicol. Chem. 35:823–835



CECs in the Chesapeake Bay and its watershed:

- Limited occurrence datasets
- Tend to mirror national studies

U.S. Environmental Protection Agency
Region III
Chesapeake Bay Program Office
Annapolis, Maryland



U.S. Geological Survey



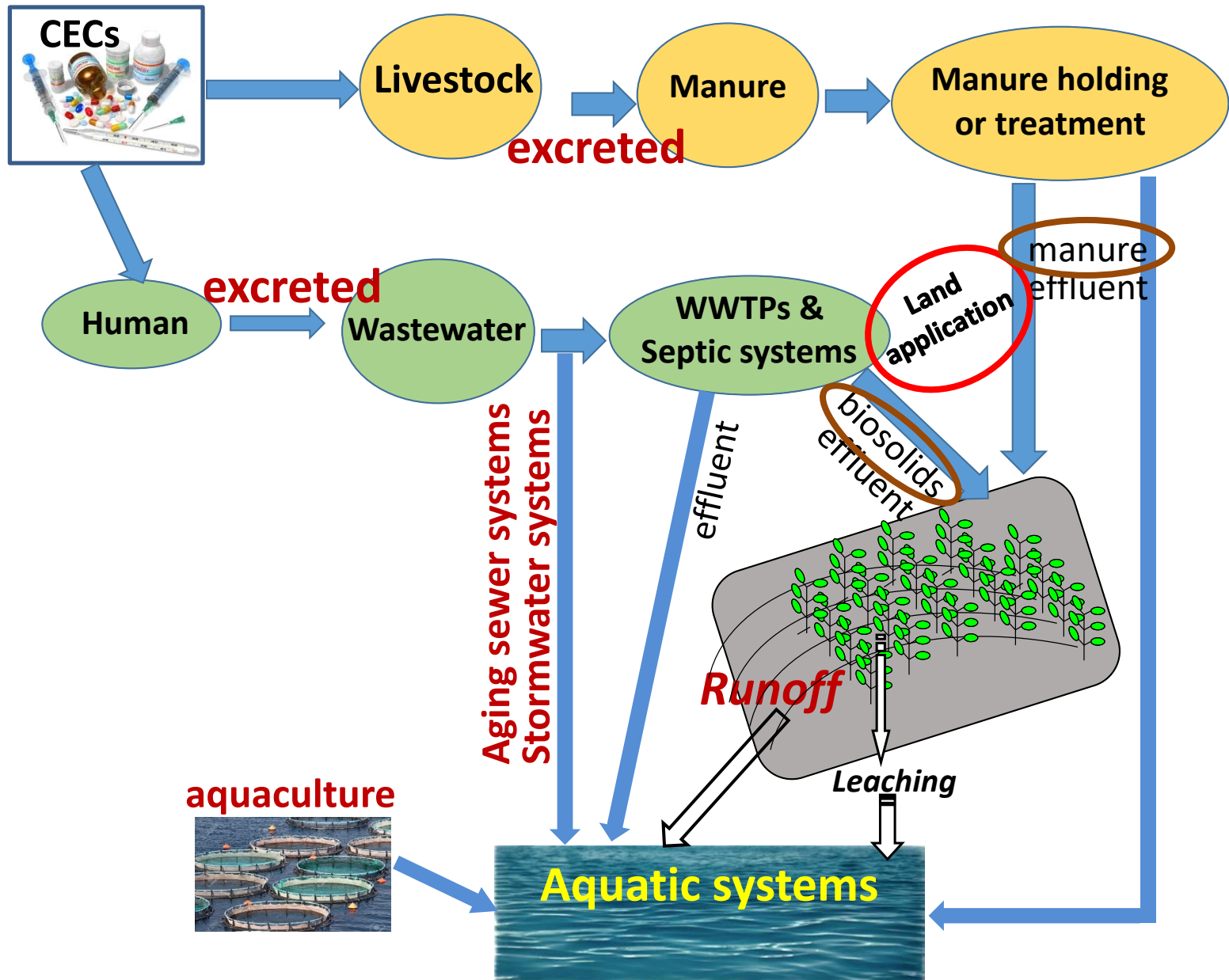
U.S. Fish and Wildlife Service
Chesapeake Bay Field Office
Annapolis, Maryland

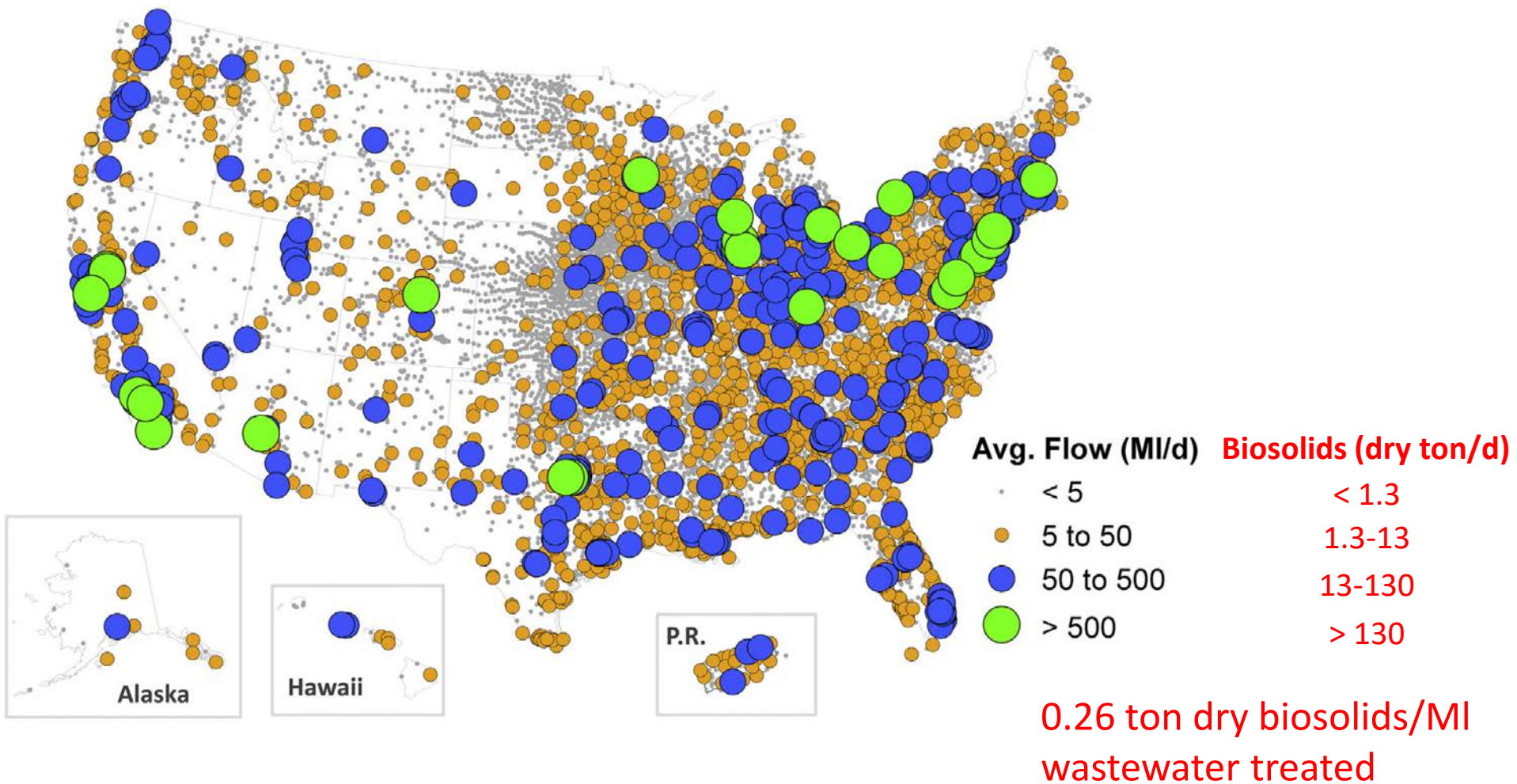


TECHNICAL REPORT | DECEMBER 2012

Toxic Contaminants in the Chesapeake Bay and its Watershed: Extent and Severity of Occurrence and Potential Biological Effects

Environmental pathways of CECs

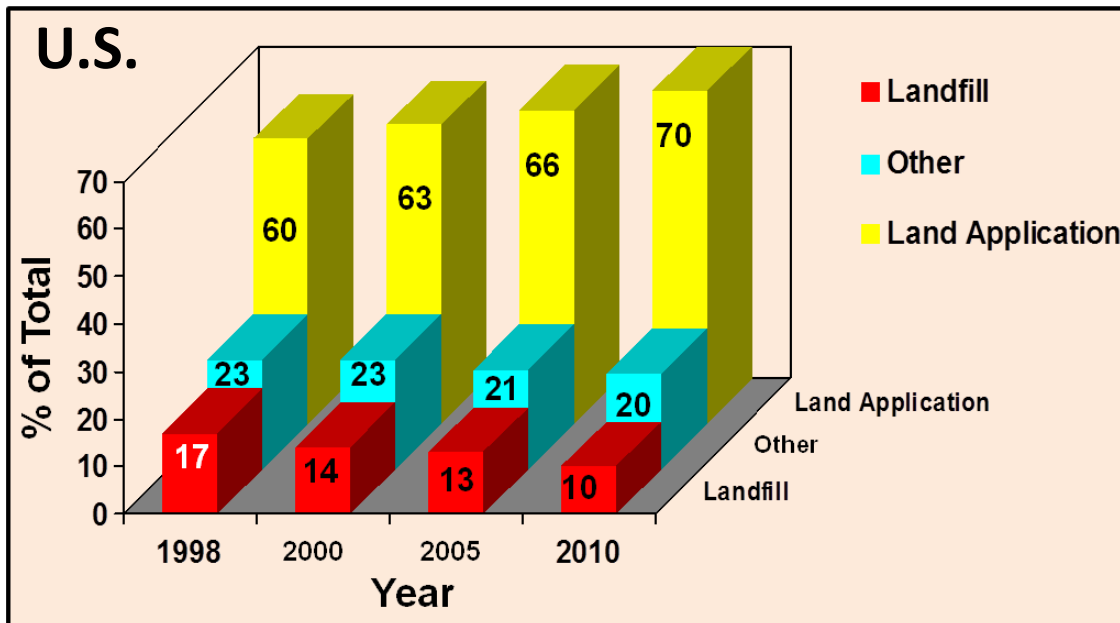
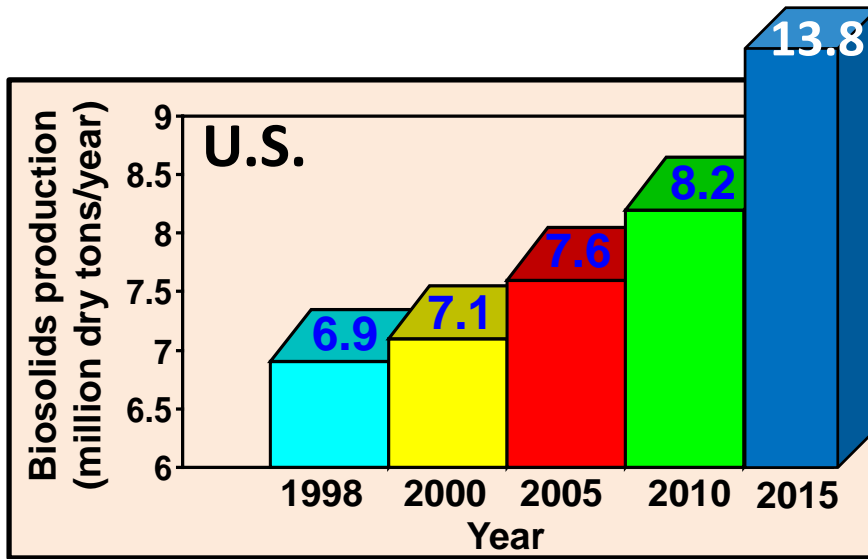




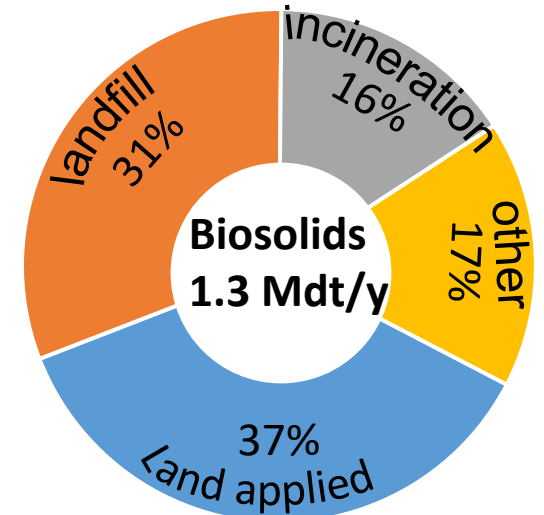
Spatial distribution of 15,014 US WWTPs classified by daily average influent flow.

Seiple et al., 2017. J. Environ. Management 197:673-680

Biosolids production, usage, and disposal



**Chesapeake Bay States
(852 WWTPs)**



**Biosolids
1.3 Mdt/y**

(0.5 M acres)

Benefits:

- primary nutrients
- secondary nutrients
- root growth promoters
- enhance soil structure
- C sequestration

(Brown et al., Environ. Sci. Technol., 2011, 45:7451)

Source: U.S. EPA, 1999

Seiple et al., 2017. J. Environ. Management 197:673-680

Occurrence of CECs in biosolids

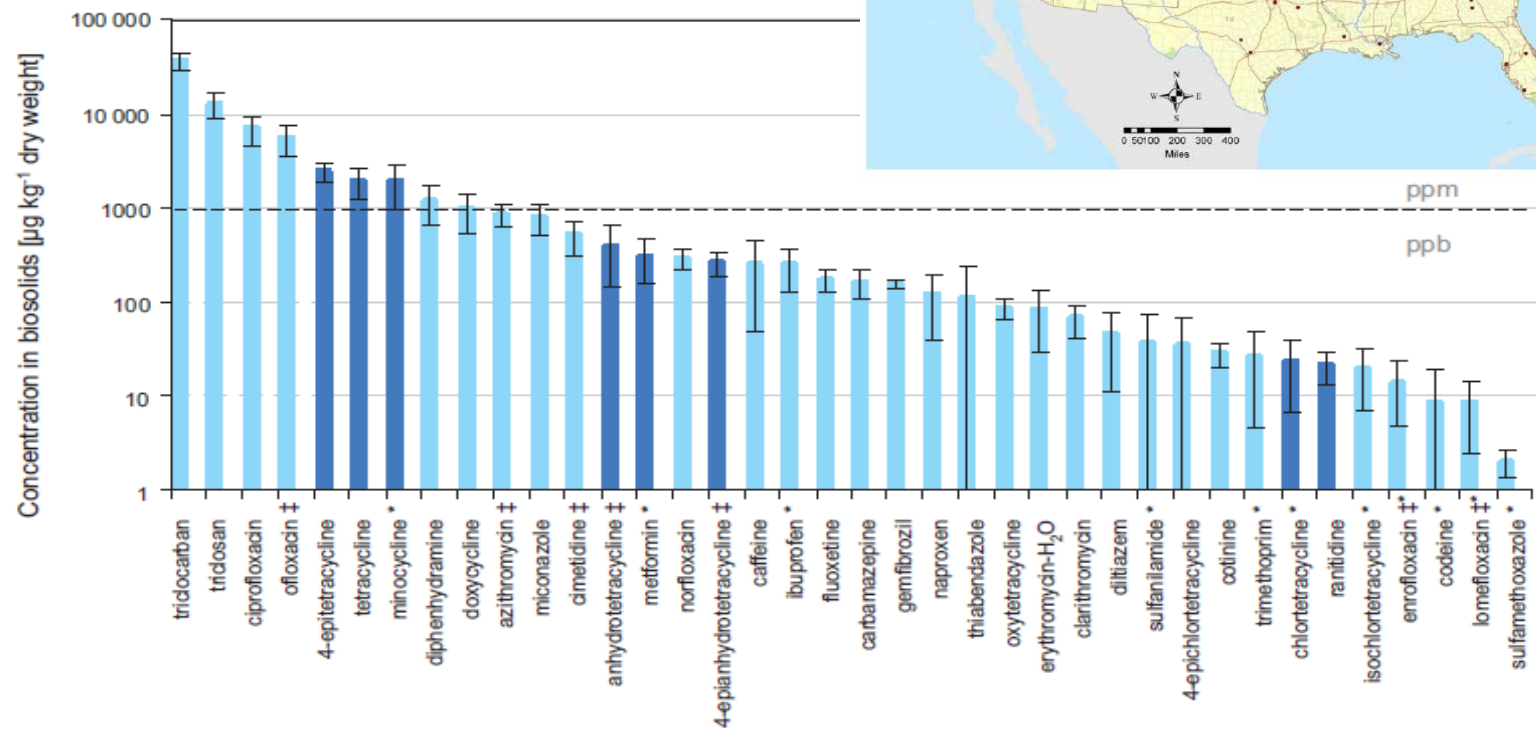


Figure 2. Geographic Distribution of 80 POTWs Originally Selected for Sampling

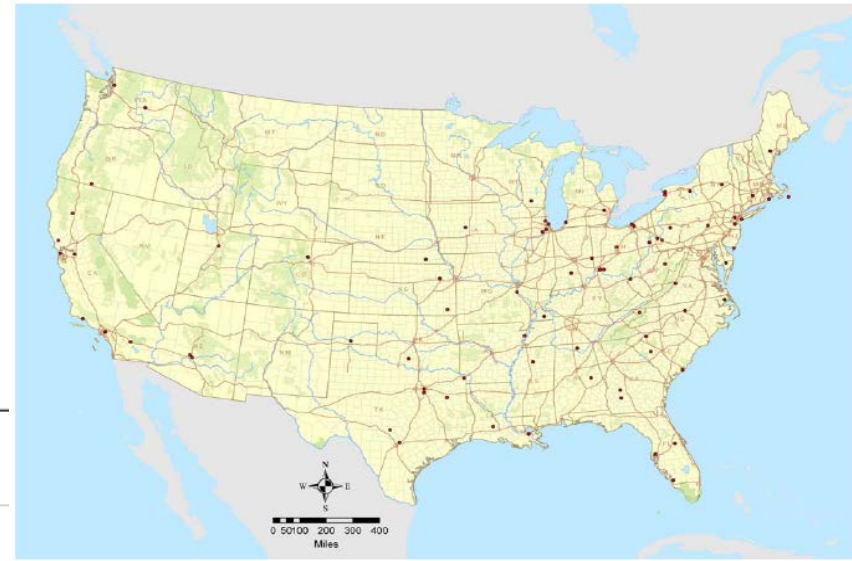


Fig. 1 – Rank order of mean concentrations for 38 PPCPs detected in composites of a total of 110 U S biosolids samples from 94 treatment plants in 32 states and the District of Columbia. Newly detected compounds are shown in darker hue. Error bars depict \pm one standard deviation ($n = 5$). Some concentrations represent estimates only (\ddagger) and some analytes were detected inconsistently (*).

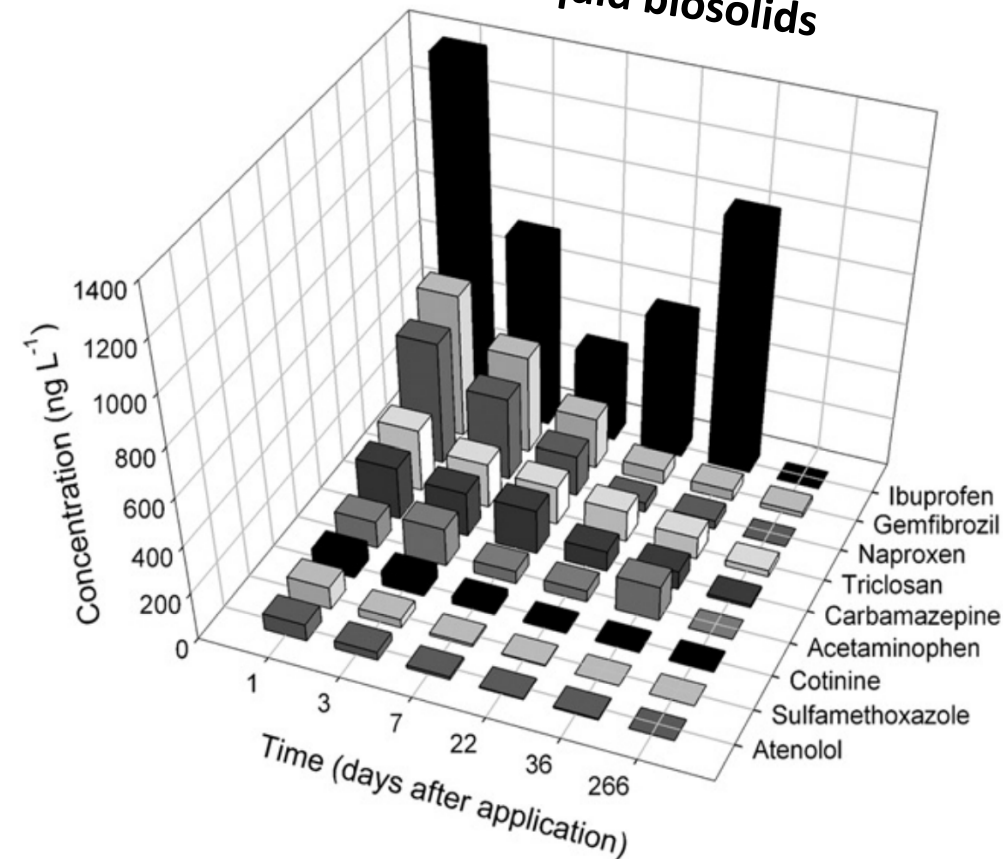
Occurrence and fate of CECs in biosolids-amended land?

TCC, TCS, PBDEs, and 4-NP in **soil after 33 consecutive years of biosolids application (Environ. Toxic. Chem. 2010. 29:597)**

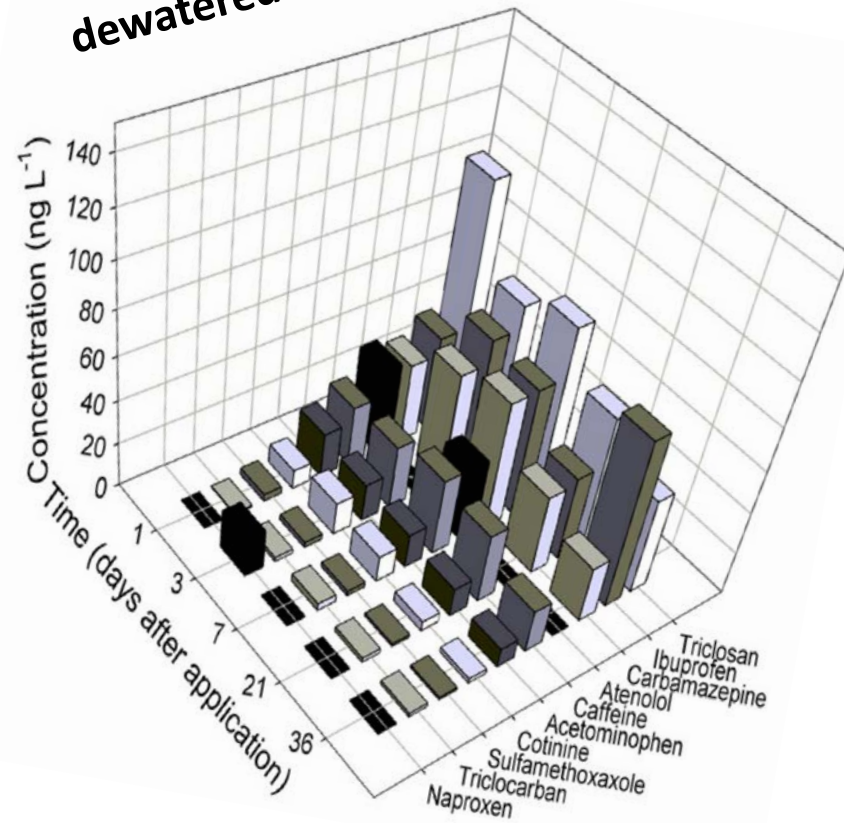
Result summary:

Target compounds	Results
Levels in soil	increase with biosolids loading sharp decrease with soil depth
PBDEs	persistent
TCC, TCS, 4-NP	less persistent
Concentrations in soil	4-NP > TCC > PBDEs > TCS (180:26:14:1)
PBDEs, 4-NP	immobile
TCC, TCS	limited mobility
Plant (corn) uptake	No

liquid biosolids



dewatered biosolids

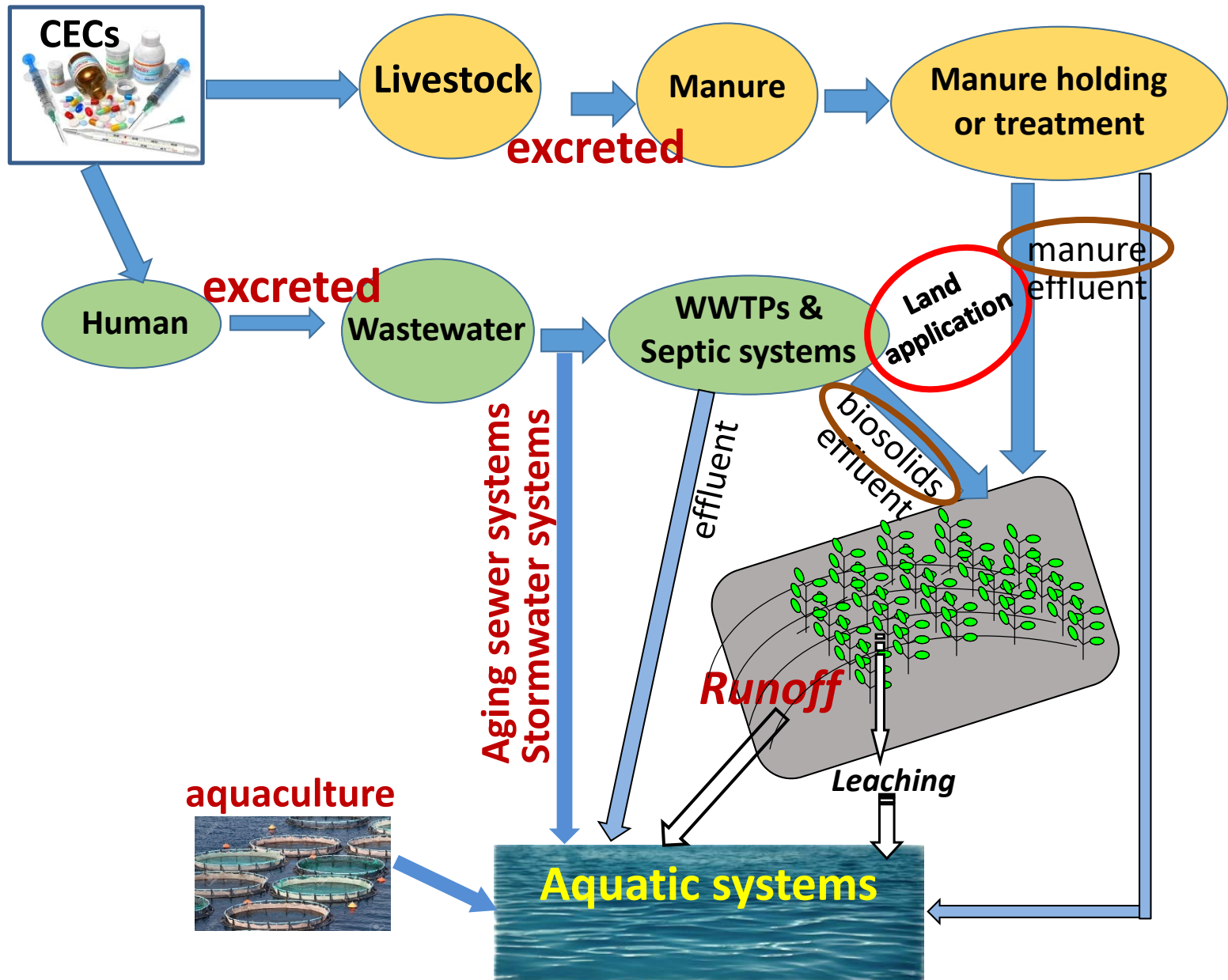


Concentrations of CECs in **surface runoff** from a field at various times post-application of broadcast incorporated municipal biosolids.

Topp et al., 2008. Sci. Total Environ. 396: 52-59

Sabourin et al., 2009, Sci. Total Environ. 407: 4596-4604

Environmental pathways of CECs



Animal manure production and usage

Exhibit 5-4: U.S. Livestock Production

Animal Type	Units	1995	2000	2005	2010	2015	2020
Dairy Cattle	Billion lbs milk/yr	156	166	178	185	193	201
Beef Cattle	Billion lbs/yr	28	28	28	29	30	30
Swine	Billion lbs/yr	19	19	21	22	23	24
Poultry	Billion lbs/yr	5	5	5	5	5	5
Sheep	1,000 head	8,886	7,998	7,998	7,977	7,939	7,872
Goats	1,000 head	2,495	2,495	2,495	2,495	2,495	2,495
Horses	1,000 head	6,000	6,325	6,642	6,970	7,314	7,661

Source: 1995-2005 values are based on USDA, 1996; 2010-2020 are values from extrapolation analysis.

- **Animal manure generated/year:**

U.S. > 1 billion tons of dry matter

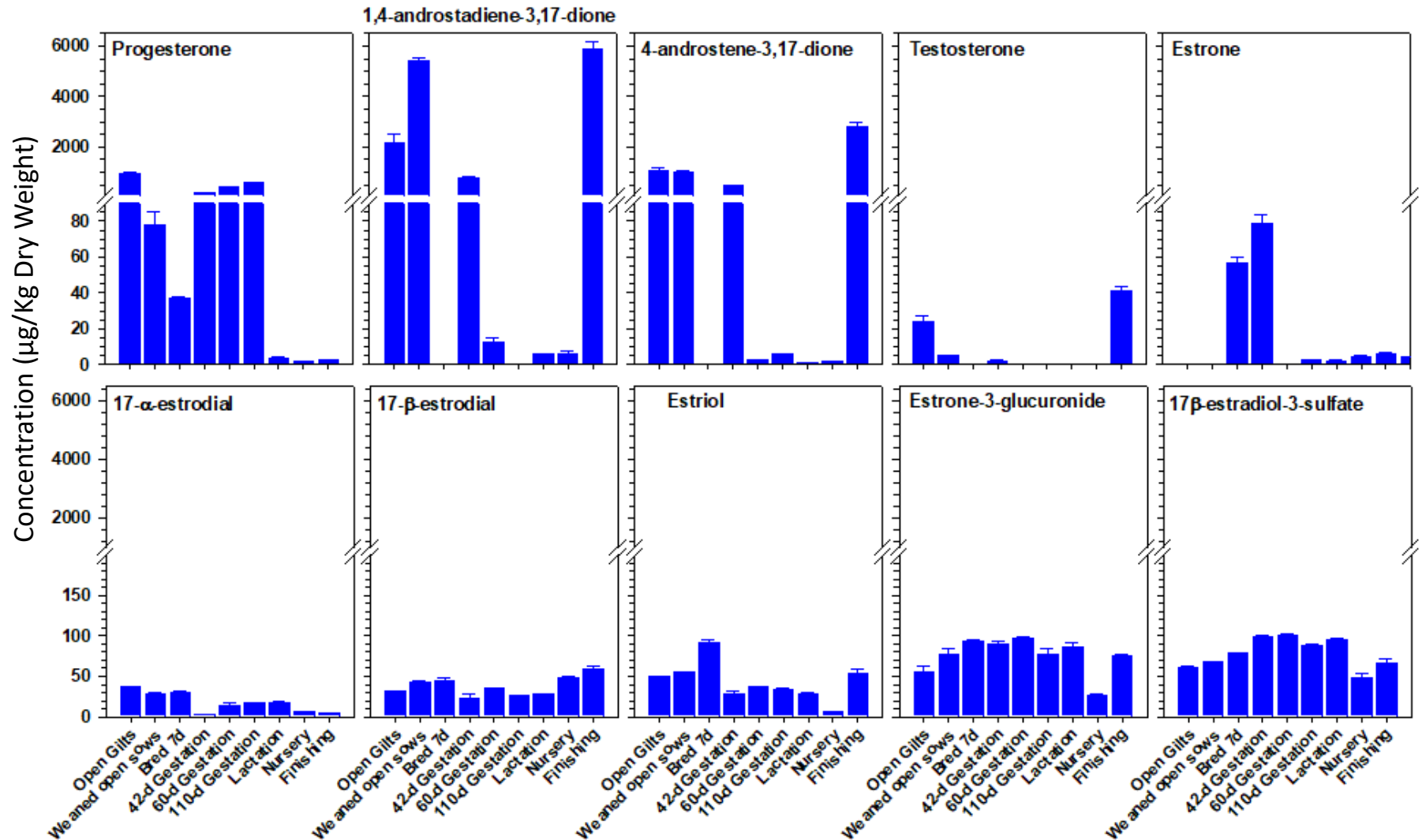
Chesapeake Bay States > 44 million tons of dry matter

- **Land application of animal manure**
**Manure is applied to 48% of farmland
in the Chesapeake Bay States**

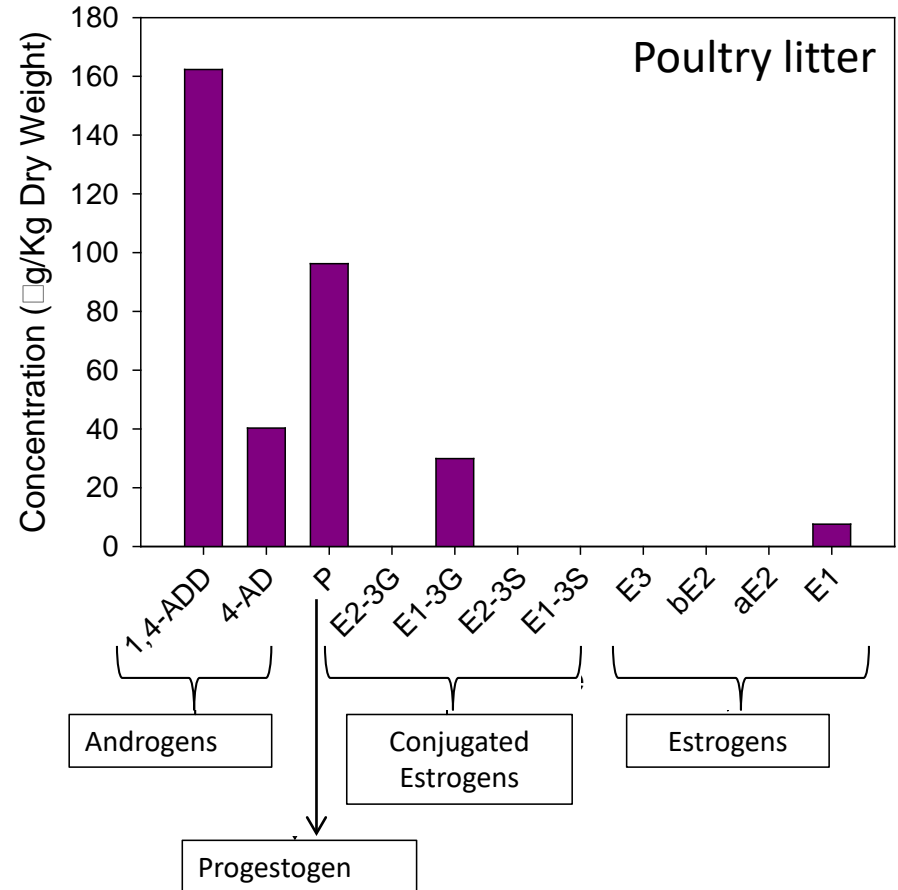
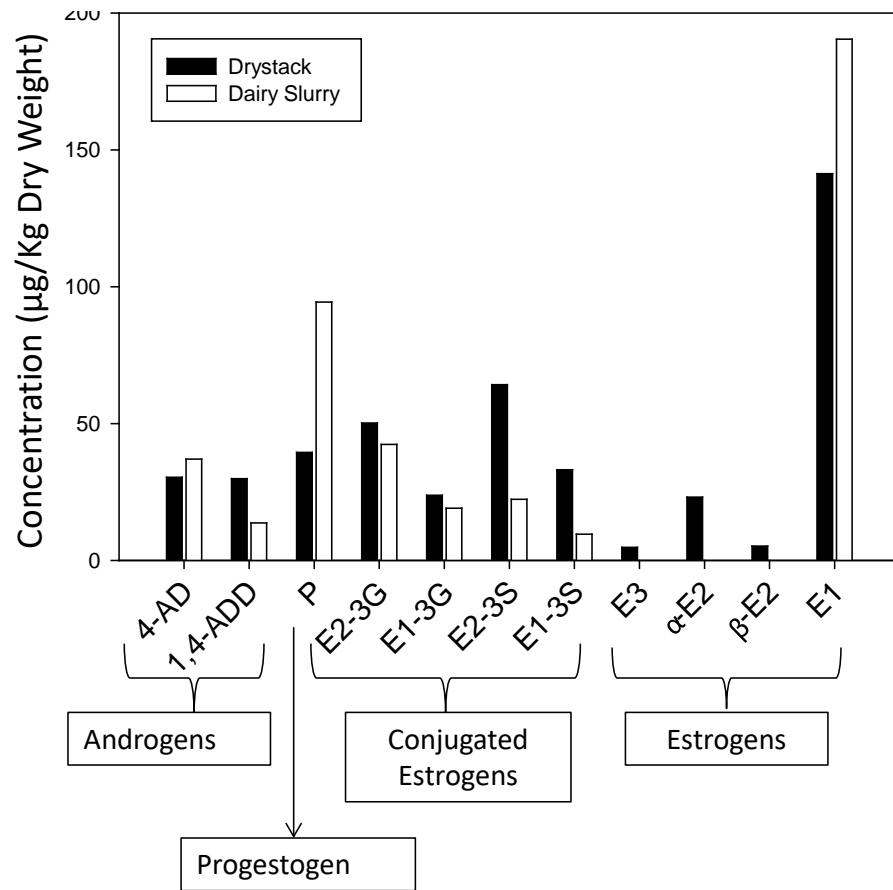
Benefits:

- primary nutrients
- secondary nutrients
- enhance soil structure
- Soil organic matter
- C sequestration

Occurrence of CECs in animal manure - hormones



*Concentrations of progestagens, androgens, and estrogens in **swine** manure generated at different life stage.*



Each year United States farm animals excrete:

Estrogens	49 t
Progestogen	4.4 t
Androgens	279 t

Lange et al., 2002, Analytic Chimica Acta. 473:27-37

Occurrence of CECs in animal manure - antibiotics

Antibiotics approved for domestic use in food-producing animals in the United States in 2015

Drug Class	Annual Totals (kg) ²	% Subtotal	% Grand Total
<i>Aminoglycosides</i> ¹	344,120	2%	2%
<i>Amphenicols</i>	44,968	<1%	<1%
<i>Cephalosporins</i> ¹	32,341	<1%	<1%
<i>Fluoroquinolones</i>	20,063	<1%	<1%
<i>Ionophores</i>	4,740,615	30%	30%
<i>Lincosamides</i> ¹	182,543	1%	1%
<i>Macrolides</i>	627,770	4%	4%
<i>Penicillins</i> ¹	936,669	6%	6%
<i>Sulfas</i>	380,186	2%	2%
<i>Tetracyclines</i> ¹	6,880,465	44%	44%
<i>NIR</i> ^{1,4}	1,387,236	9%	9%
Subtotal	15,576,975	100%	100%

(FDA Annual Summary Report on Antimicrobials Sold or Distributed in 2015 for Use in Food-Producing Animals)

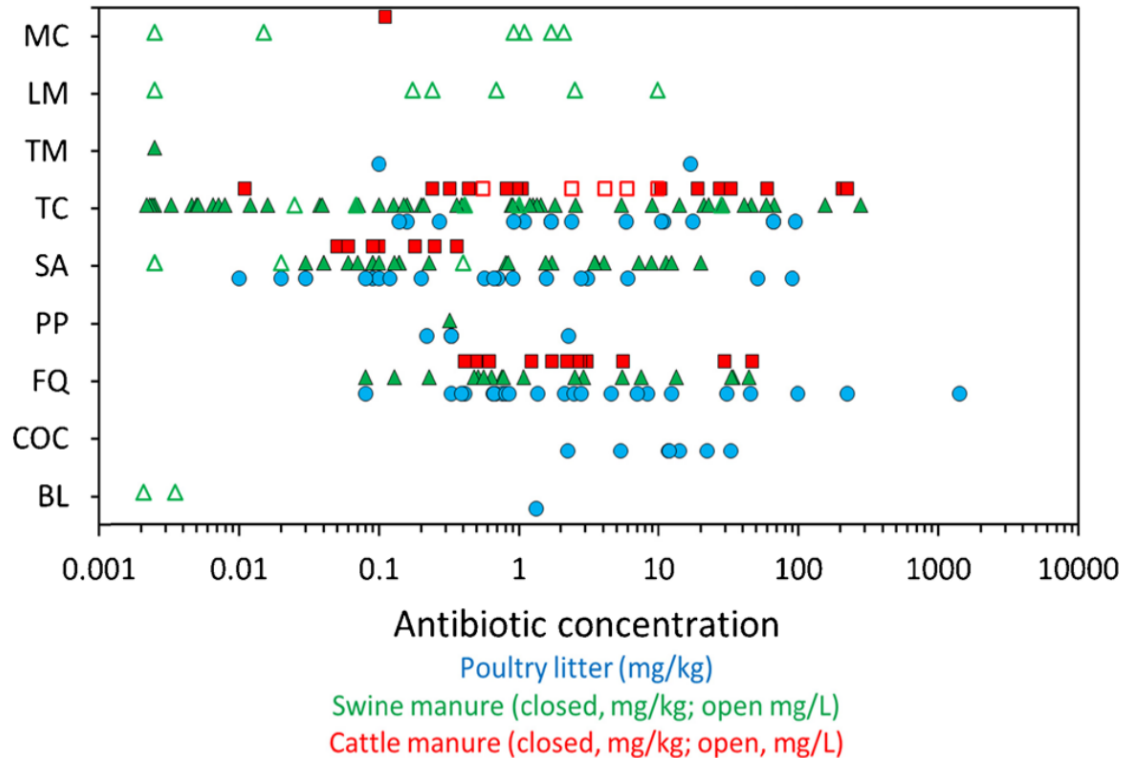


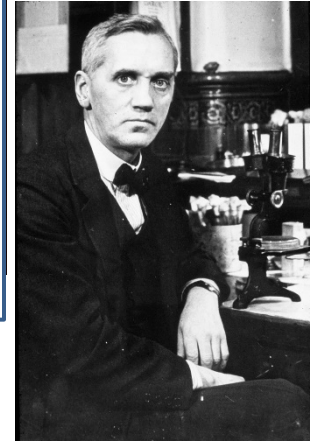
Fig. 1 Antibiotic concentrations detected in poultry, swine, and beef cattle manure. Data was aggregated from available reports [13, 16, 18, 21, 39–59]. Antibiotic class codes on the y-axis are as follows: *MC* macrolide, *LM* lincosamide, *TM* trimethoprim, *TC* tetracycline, *SA* sulfonamide, *PP* polypeptide, *FQ* fluoroquinolone, *COC* coccidiostat, *BL* beta-lactam. For clarity, only the minimum and maximum antibiotic concentrations from individual studies were included here. This list is not exhaustive but is meant to convey the relative antibiotic detection and concentration ranges in animal manures

(Van Epps & Blaney, 2016, *Curr Pollution Rep.* 2:135–155)

Why are antibiotics used in animal production?

Alexander Fleming' Nobel lecture in 1945:

*"The time may come when penicillin can be bought by anyone in the shops. Then there is the danger that the ignorant man may easily underdose himself and by exposing his microbes to non-lethal quantities of the drug make them **resistant**."*



Alexander Fleming

1955 – 1st scientific report on usage of antibiotics in food animal production for **growth promotion** (*Science*. 1955. 121:733-734)

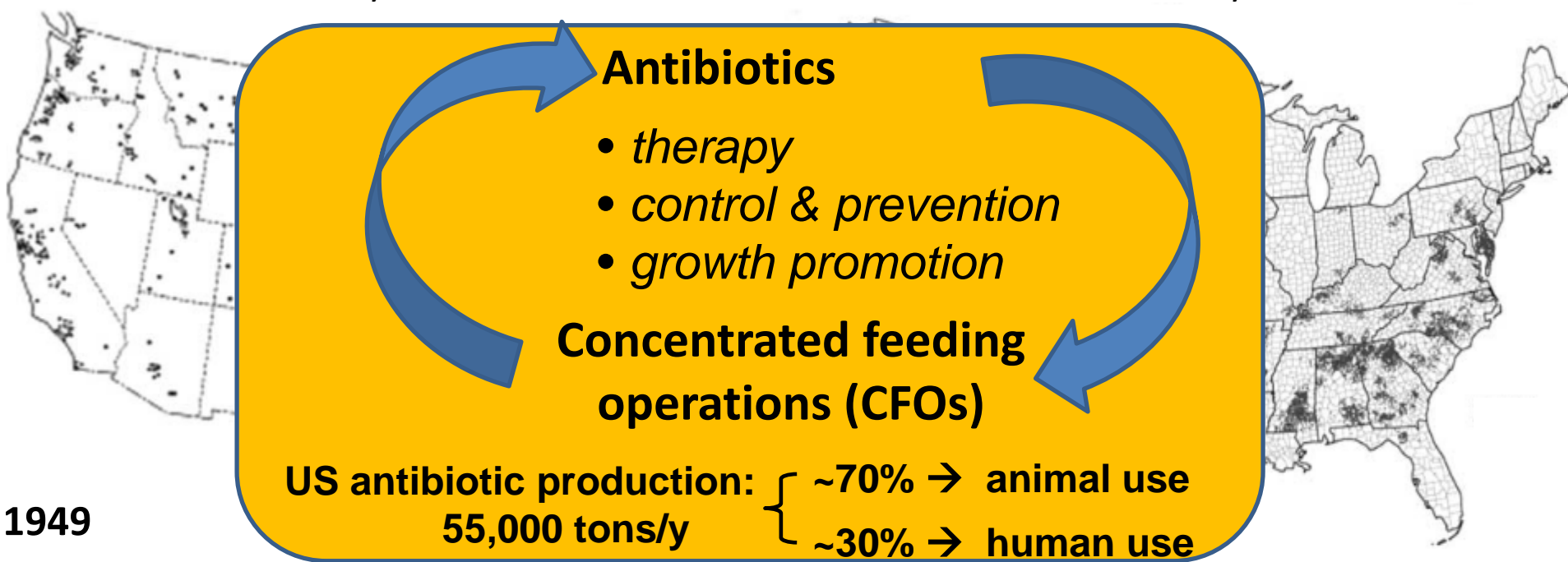
From the practical point of view, this study indicates that antibiotics and arsonic acids would be of greatest value in promoting growth (i) when first used in a poultry-feeding enterprise and (ii) under the environmental conditions usually surrounding farm feeding. If once used and then discontinued, feeding performance may still be somewhat improved, because the contamination level has been lowered. Continued use of these substances at low levels, however, would help to insure maximum results, because an "infected" area can be reestablished through disuse (5). Indications are that such use of antibiotics and arsonic acids in feeds will not build up a resistant microflora that future poultry populations will have difficulty to live with.

- 1950-now,
US animal production ↑ 100%
of production units ↓ 80%



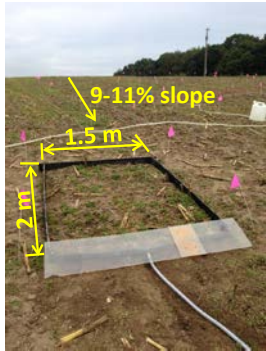
● >50,000 chickens /year

● >1,000,000 chickens/year



Journal of Water and Health. 2010. 8:646-670.

Occurrence and fate of CECs in manure-amended land?



2x1.5 m²
plot



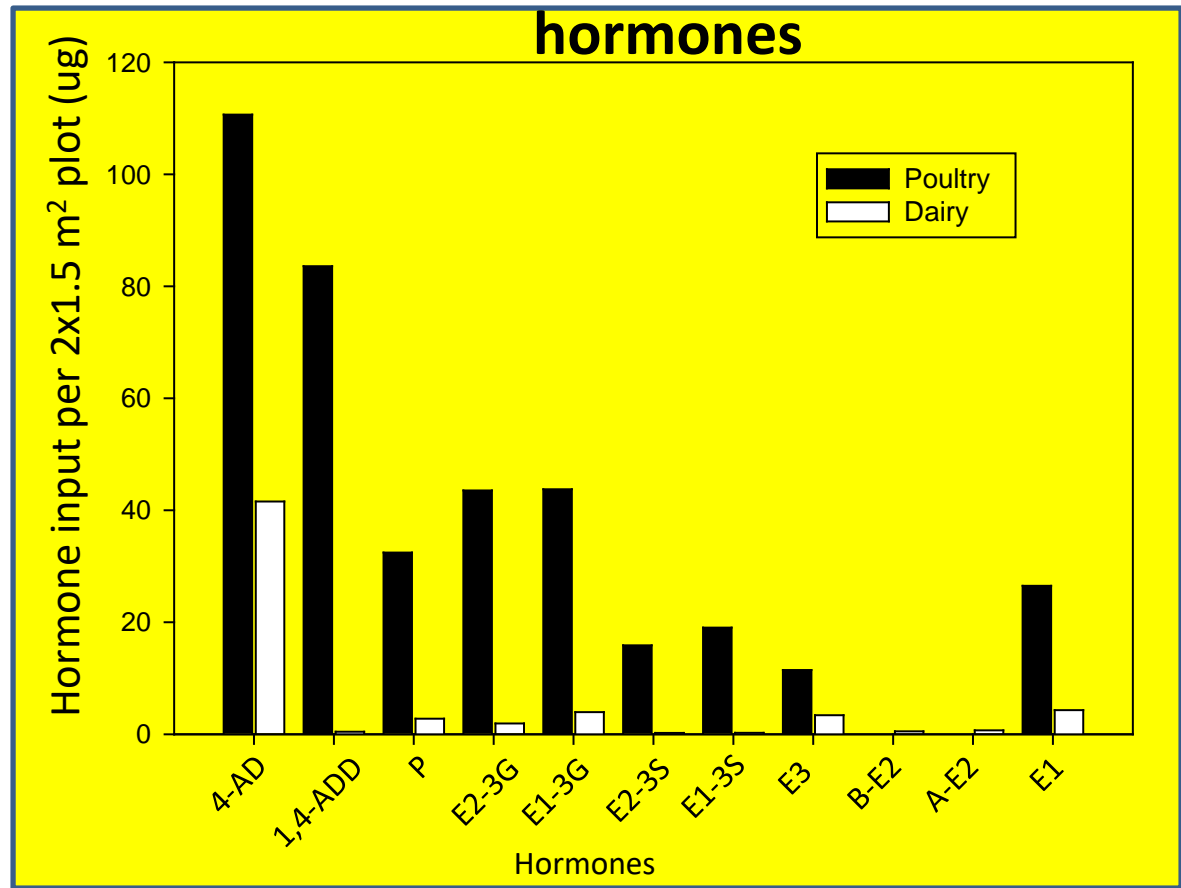
Surface
application



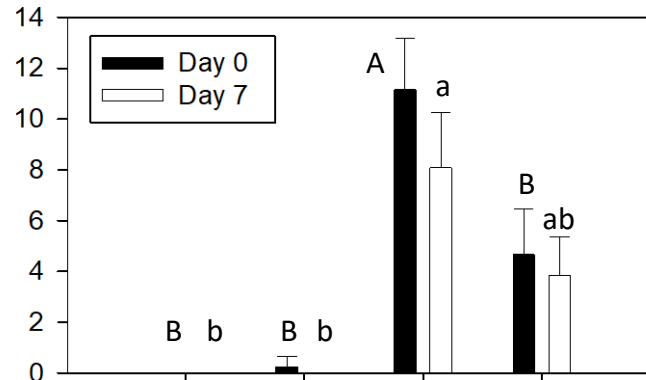
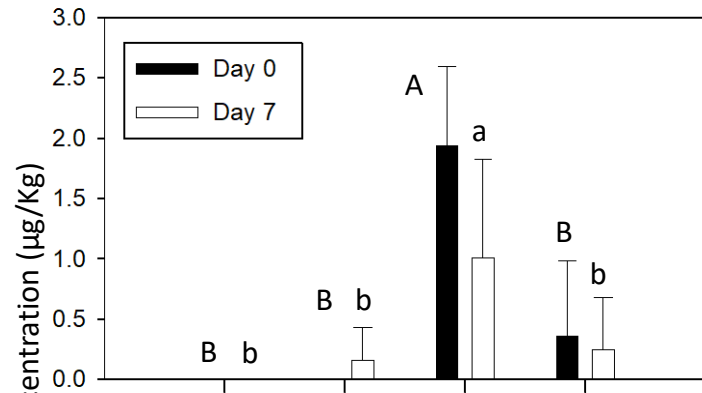
Subsurface
injection



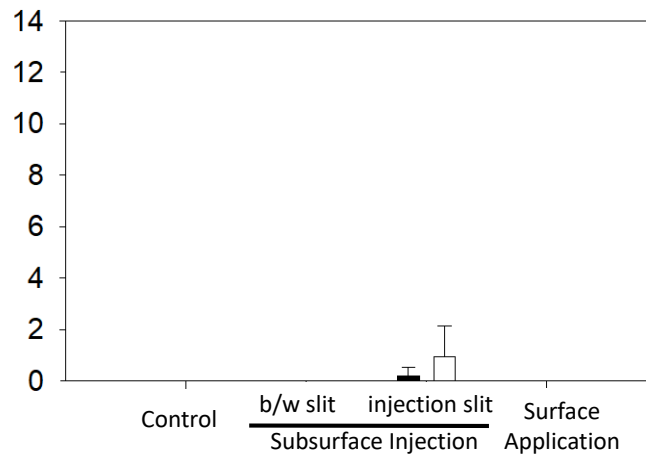
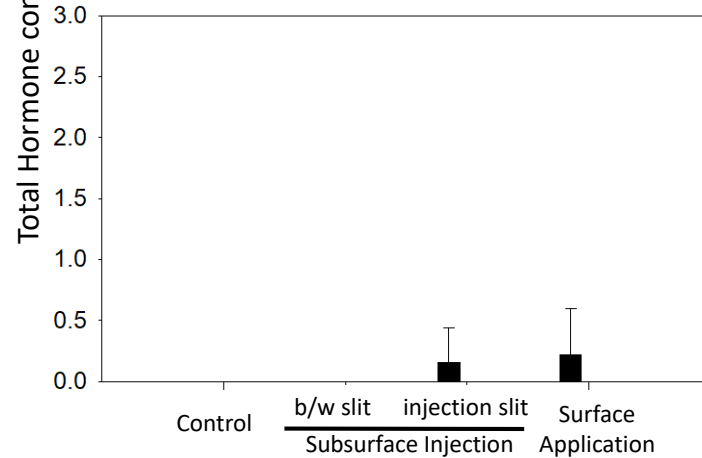
Rainfall
simulation



Total hormone concentration in soil



0-5 cm

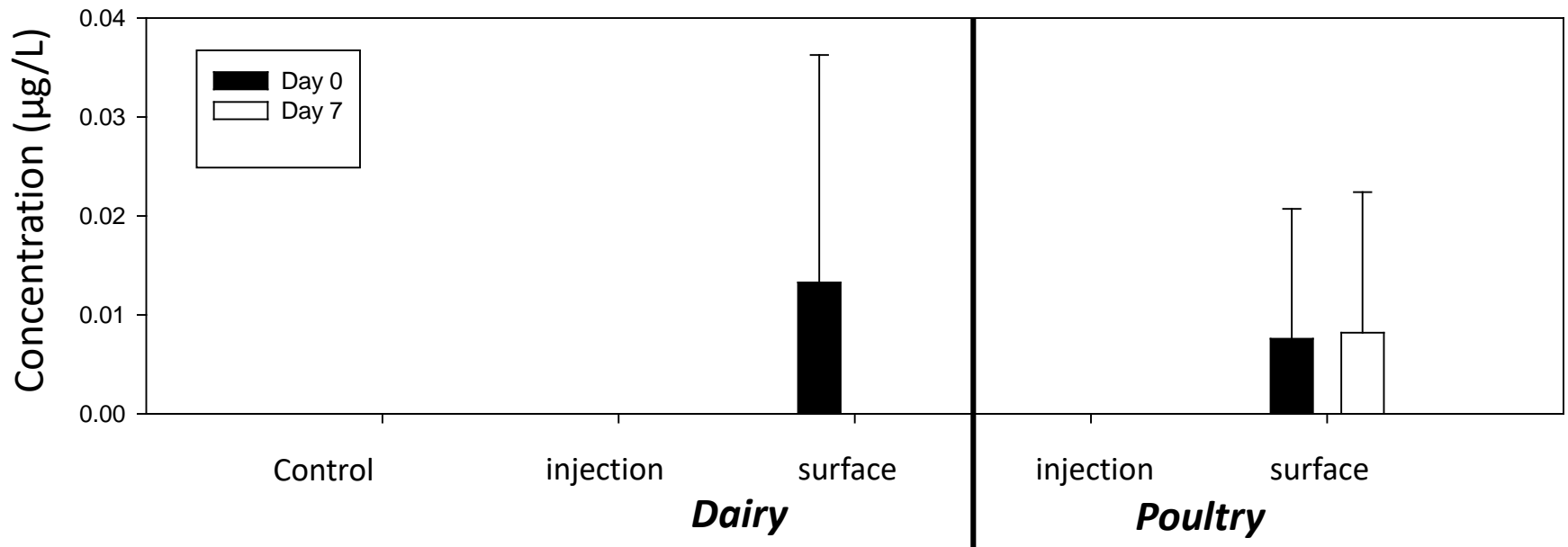


5-20 cm

- **Minimal leaching** – concentrated in top 0-5 cm soil

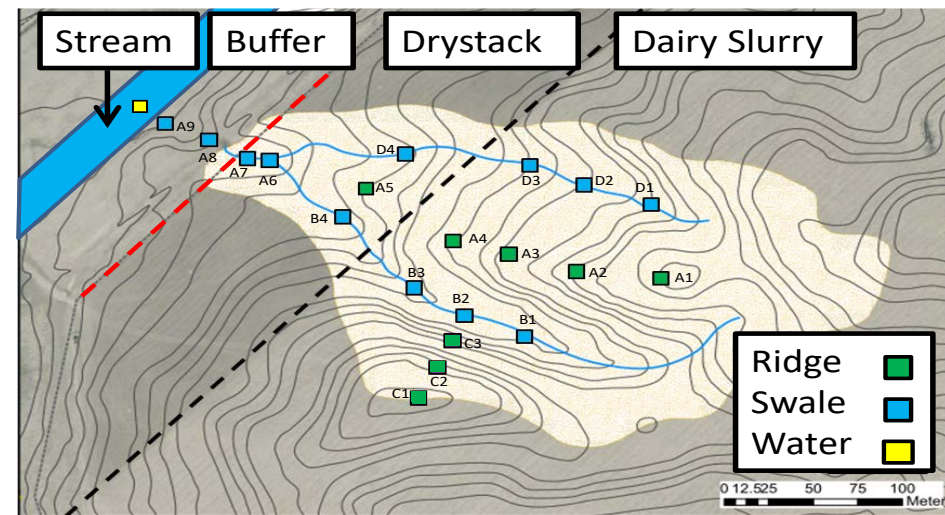
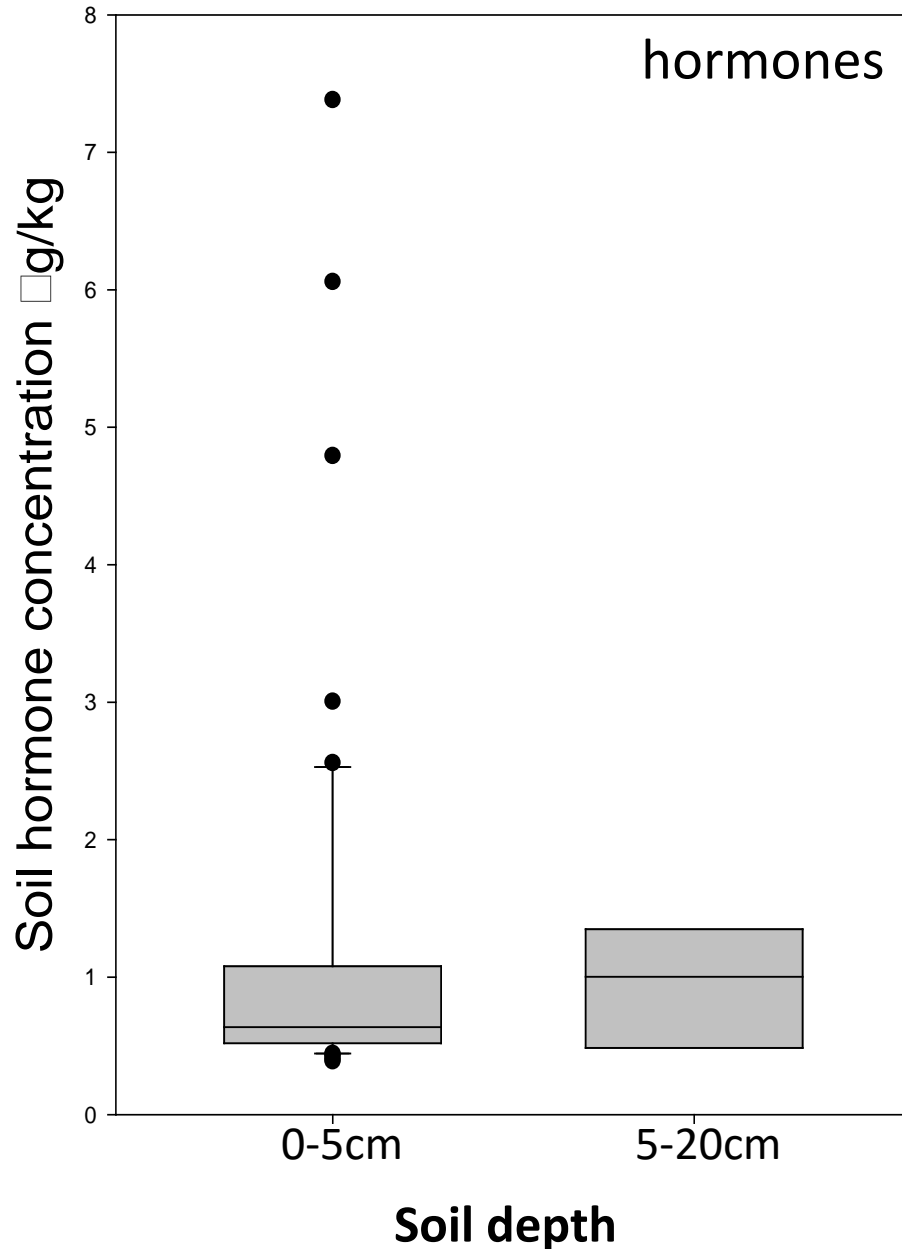
- **Concentrated in the injection slits**

Total hormone concentration in surface runoff



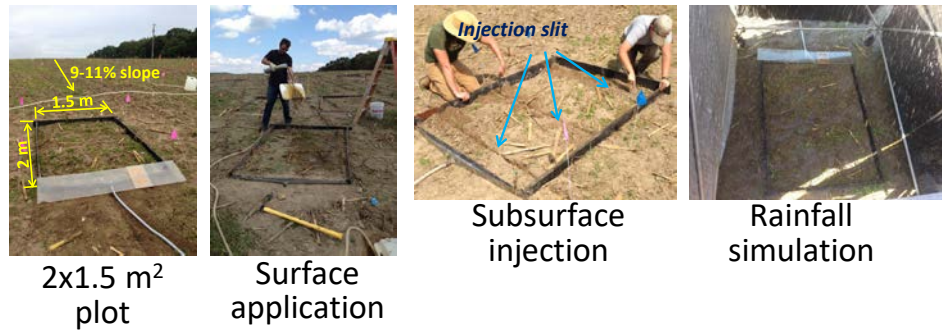
- ***Subsurface injection reduces hormone surface runoff***

Surface applied manure

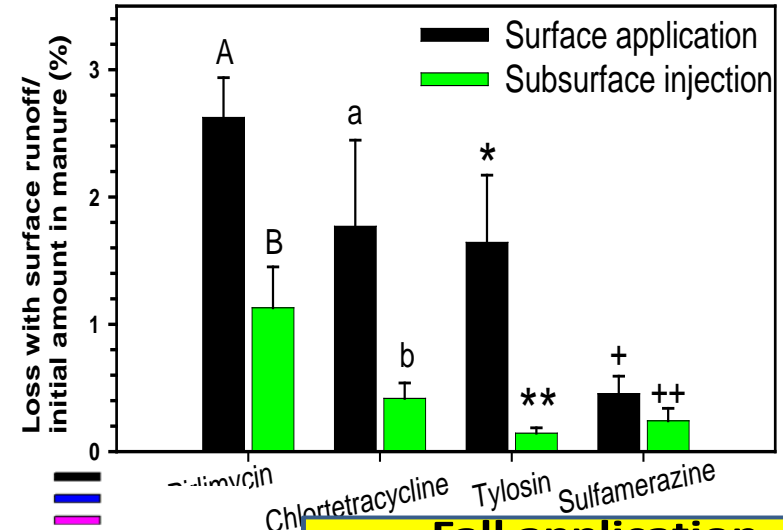


- **Minimal leaching** – concentrated in top 0-5 cm soil
- **Downslope movement**
- **Buffer is effective** – none-detectable in the stream
- **Progesterone more persistent** – detectable 3 months after manure application
- **Similar observation** – DE coastal plain poultry litter amended soils (Water Air Soil Pollut, 2012, 223:2821–2836)

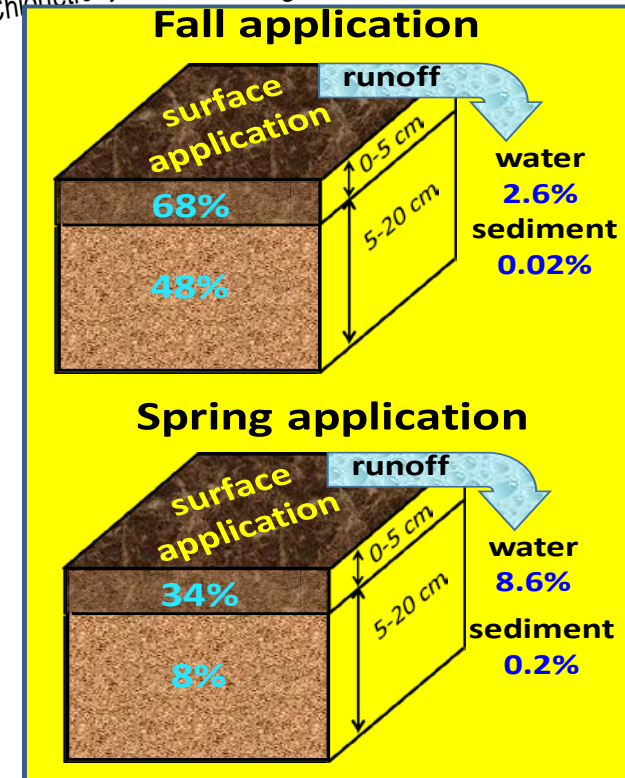
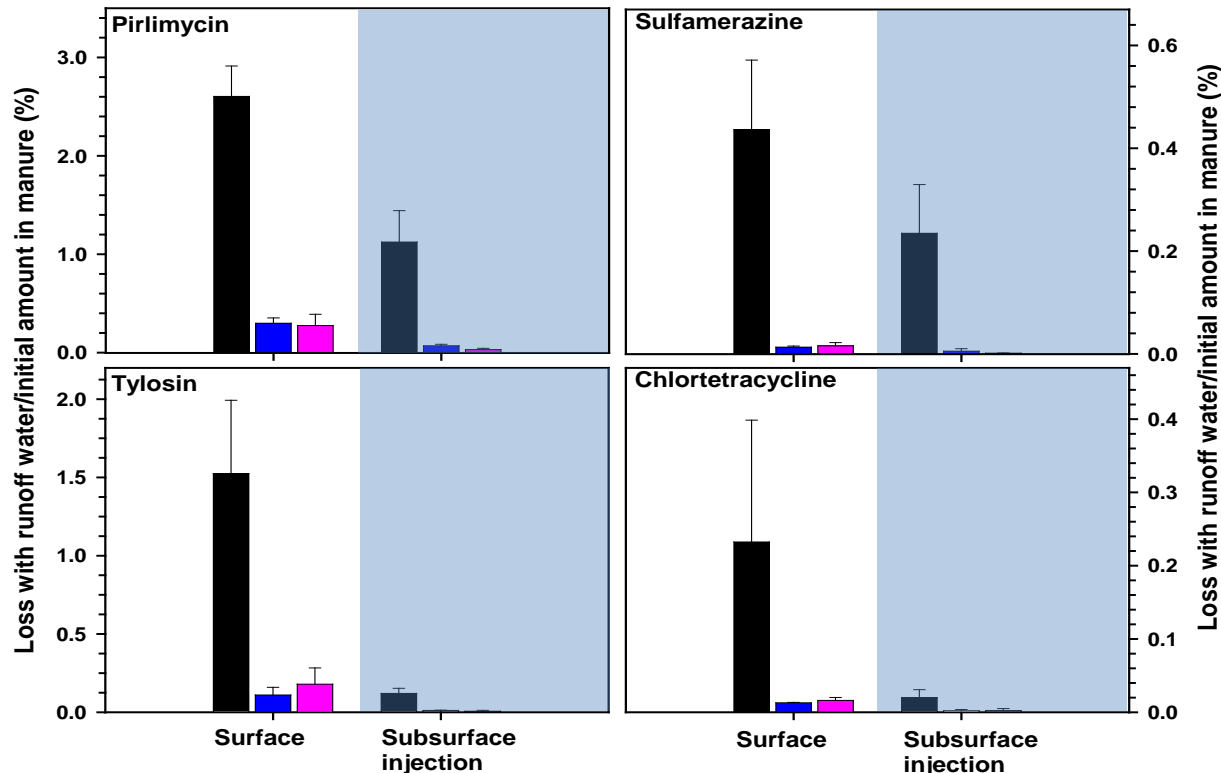
Occurrence and fate of CECs in manure-amended land?



Antibiotics in surface runoff

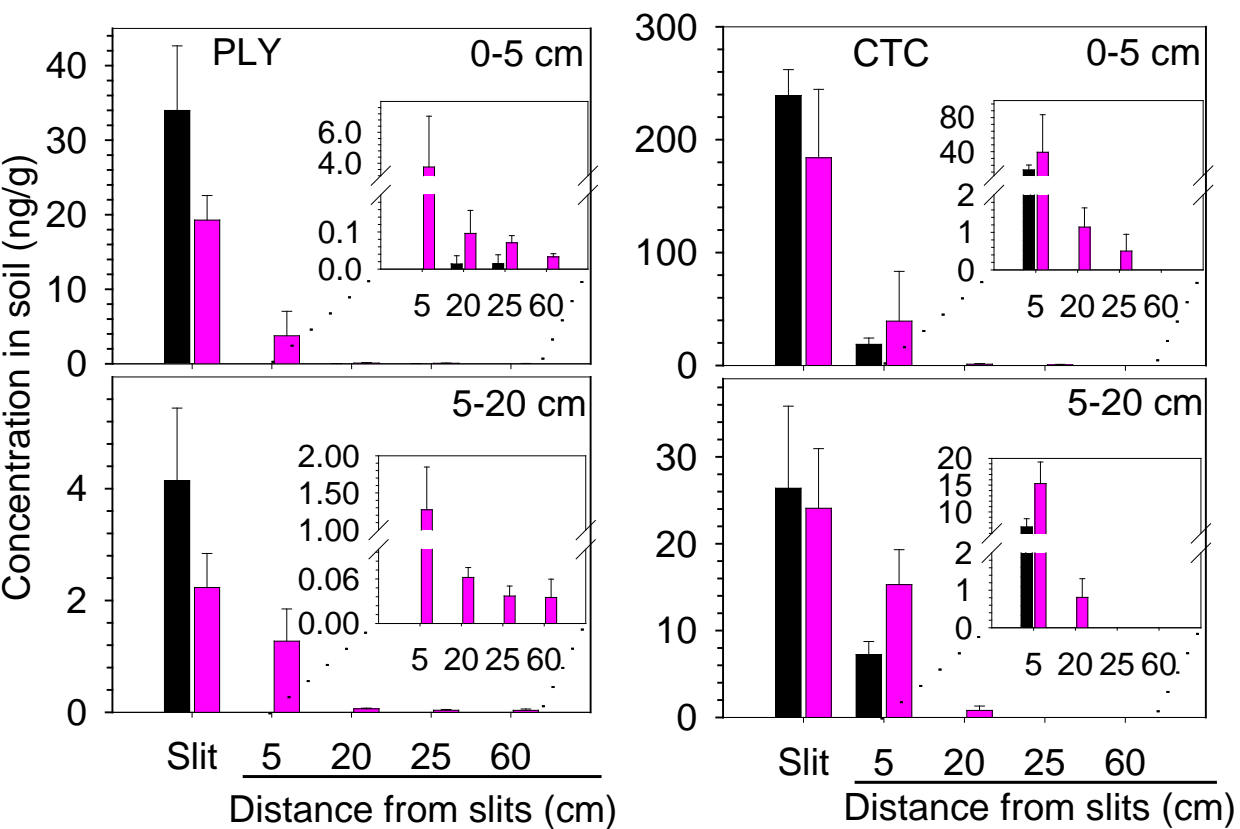


Time gap b

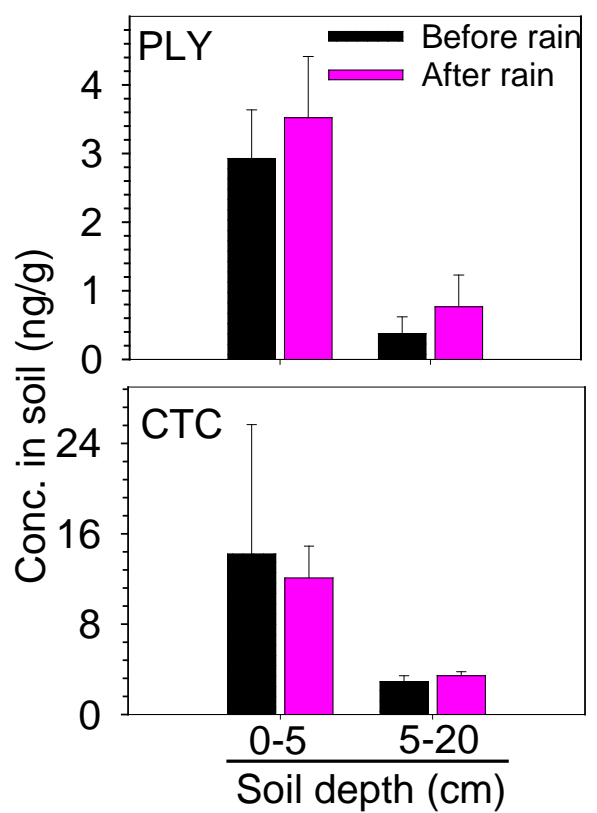


Spatial distribution of antibiotics in soil

Subsurface injection
(rain on day 3)

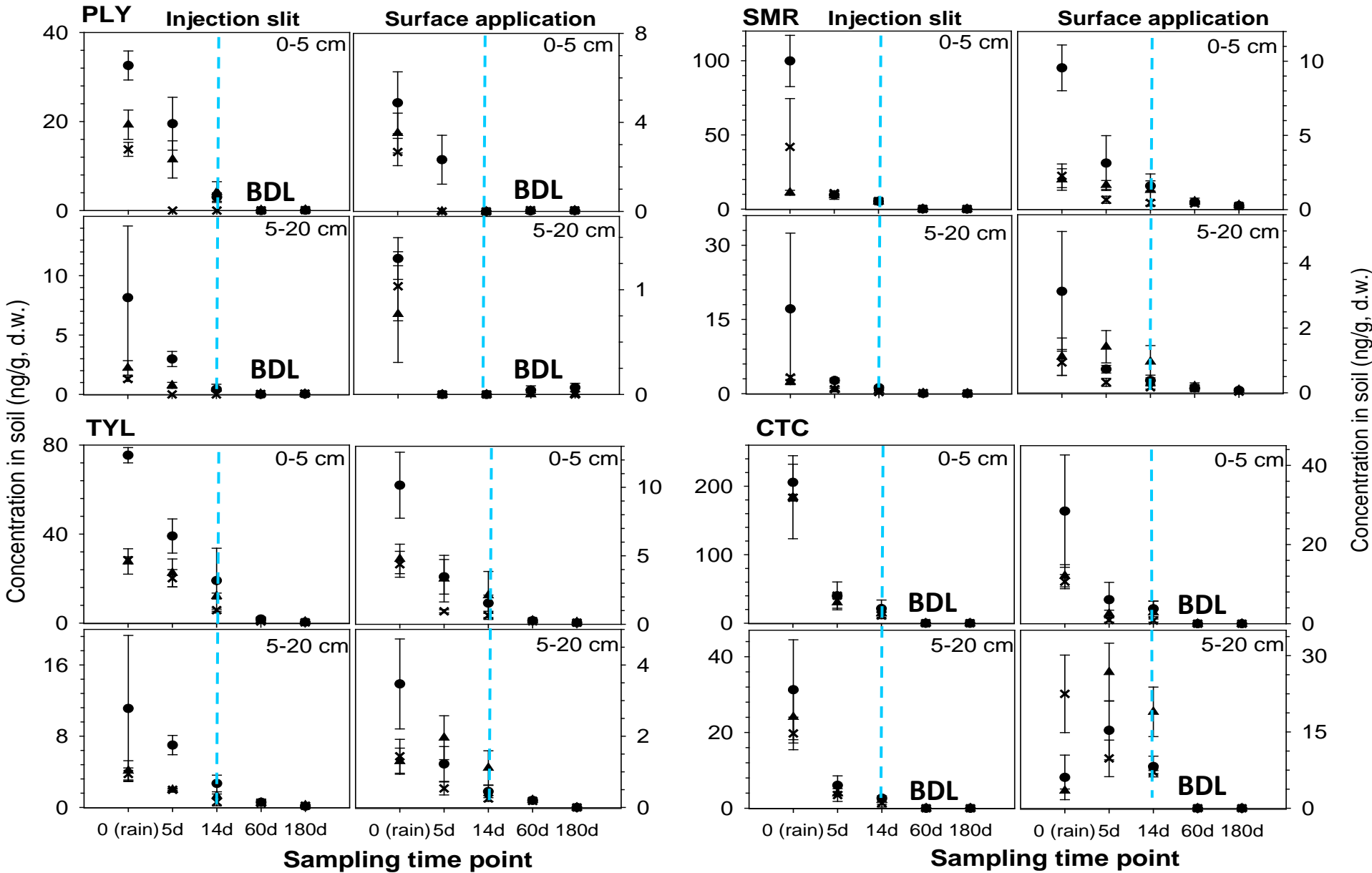


Surface application
(rain on day 3)



Temporal change of antibiotics in soil

Days between manure application & rain event: ● 0 ▲ 3 × 7



Chemicals (Contaminants) of Emerging Concern from Rural Nonpoint Sources

- **We do know:**

- Wide spread occurrence of CECs in biosolids and manure
- Land application of biosolids and manure → occurrence of CECs in soil and runoff → aquatic environment
- Proper biosolids/manure management and land application methods can reduce input of CECs

- **What we don't know:**

- CECs budget in CBW?
- Antibiotic resistant?
- Environmental impact?

The screenshot displays the website for the Mid-Atlantic Water Program, a partnership of USDA CSREES and Land Grant Colleges and Universities. The page is titled "Nutrient Budgets for the Mid-Atlantic States" and includes a navigation menu on the left with links to "Regional Program", "Introduction", "Glossary", "Contact Us", "Budgets", and "Budget Details". The "Budgets" section is currently selected, showing links for Delaware, Maryland, Pennsylvania, Virginia, West Virginia, and Mid-Atlantic. The "Budget Details" section includes links for "Data Sources", "Methods", "Assumptions", "Limitations", and "References". The main content area explains that the program provides county- and state-level nitrogen and phosphorus nutrient budgets for agricultural cropland in Delaware, Maryland, Pennsylvania, Virginia, and West Virginia. It notes that these budgets aim to improve water quality protection by addressing the lack of balance between available nutrient supplies and nutrient use by crops. The page also mentions funding from the US Department of Agriculture and the Chesapeake Research Consortium, and lists the Mid-Atlantic States: Delaware, Maryland, Pennsylvania, Virginia, West Virginia, and Mid-Atlantic. Logos for the University of Delaware, University of Maryland, Penn State, Virginia Tech, and West Virginia University are visible at the bottom.

Applying knowledge to improve water quality

Mid-Atlantic Water Program
A Partnership of USDA CSREES & Land Grant Colleges and Universities

Nutrient Budgets for the Mid-Atlantic States

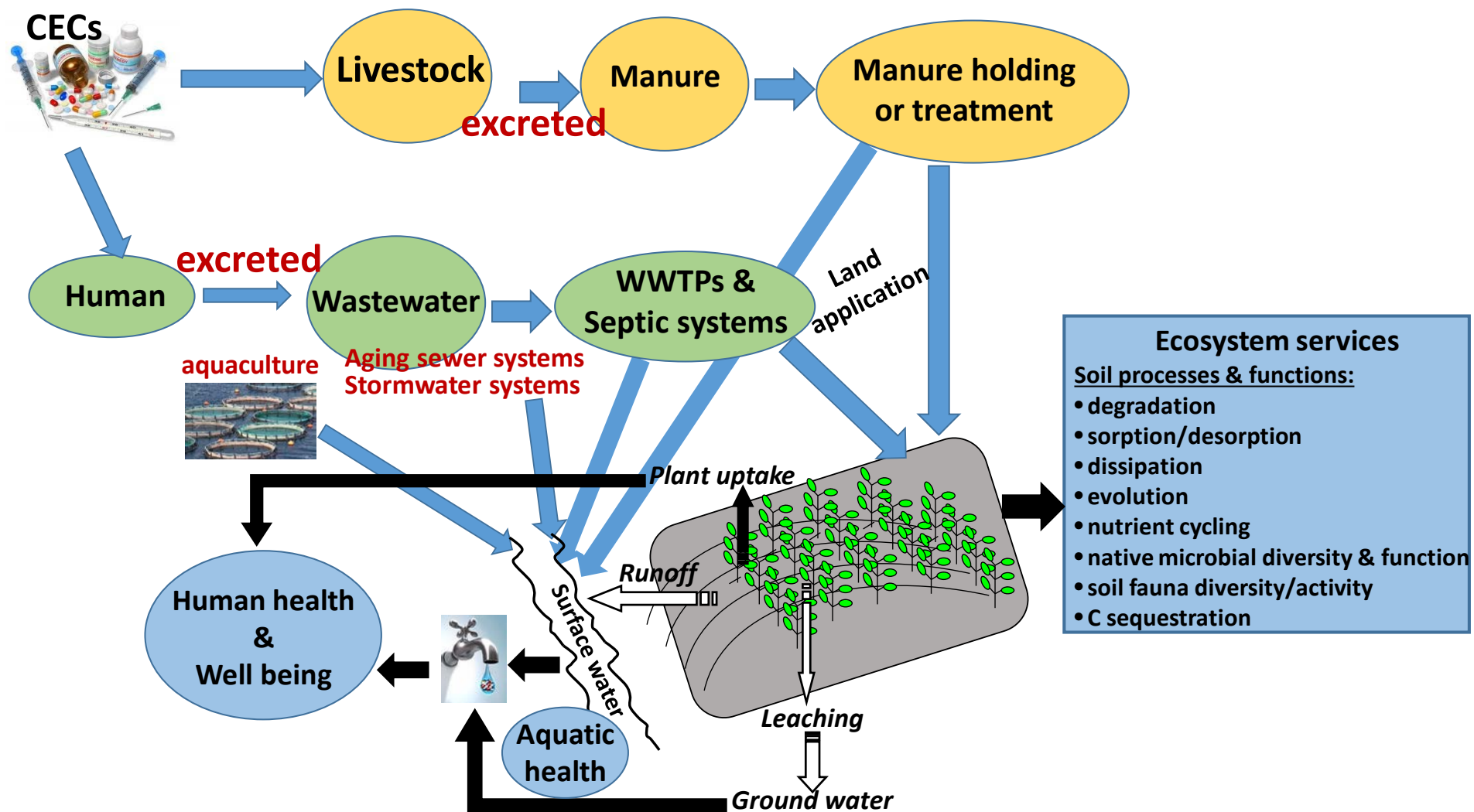
As part of the [Mid-Atlantic Water Program](#), Cooperative Extension specialists and researchers at several universities have developed county- and state-level [nitrogen](#) and [phosphorus nutrient budgets](#) for agricultural [cropland](#) in Delaware, Maryland, Pennsylvania, Virginia and West Virginia. These budgets provide the potential to improve water quality protection by supporting activities that address the lack of balance between available nutrient supplies and nutrient use by crops in a region. These nutrient budgets incorporate information updated from data sources such as the 2007 Census of Agriculture and 2007 fertilizer sales. To learn about specific cropland budgets, click on a state below.

Funding for this effort was provided by the US Department of Agriculture and the Chesapeake Research Consortium. This website is dedicated to the vision and memory of Les Lanyon, Professor of Soil Science and Management, Penn State University, and original leader of the Mid-Atlantic nutrient budget team.

Mid-Atlantic States:

- [Delaware](#)
- [Maryland](#)
- [Pennsylvania](#)
- [Virginia](#)
- [West Virginia](#)
- [Mid-Atlantic](#)

Logos at the bottom: University of Delaware, University of Maryland, Penn State, Virginia Tech, West Virginia University.



Pathways of chemicals of emerging concern and their impact

Acknowledgements

- Graduate students & postdoc researchers
- Collaborators

- USDA-NIFA award #2013-67019-21355
- USDA-NIFA award #2014-05280
- USDA-NIFA award # 2017-67019-26401
- USDA-NIFA award #2017-68003-26498

- College of Agriculture and Life Sciences, Virginia Tech

Thank you!

Questions?