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Survey of Human Use Pharmaceuticals in the Chesapeake Bay

Anthony S. Pait[1], Edward T. Furlong[2], Judd O. Nelson[3], S. Ian Hartwell[1], Percy A. Pacheco[4], Robert A. Warner[1]

[1]NOAA, National Centers for Coastal Ocean Science
[2]U.S. Geological Survey/ National Water Quality Laboratory
[3]University of Maryland
[4]NOAA, Special Projects Office

Toxics in Maryland Waters
College Park, MD
November 6, 2009

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Partners

- U.S. Geological Survey
  - National Water Quality Laboratory, Denver
- University of Maryland
  - College Park
- NOAA
  - National Centers for Coastal Ocean Science
    - National Status and Trends (NS&T) Program

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NOAA's National Status and Trends Program

- NS&T has monitored contaminants in coastal waters for over 20 years through the Mussel Watch Project
- Approximately 250 sites
- Bivalves (e.g., oysters and mussels) sampled every 2 years; sediments every 10 years
- Organic and inorganic contaminants (150+)
  - PAHs (59)
  - PCBs (38)
  - Chlorinated pesticides (31)
  - Major and trace elements (16)
  - PBDEs (38 congeners) currently being added
- Emerging contaminants

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National Status and Trends Program: Emerging Contaminants

- Emerging Contaminants - previously unknown or unidentified classes of compounds, such as pharmaceuticals, which have the potential for negative effects in the environment
- Ongoing effort within the NS&T Program to assess the presence of emerging contaminants in estuarine and coastal waters
- Assess the distribution and concentration; ultimately determine if any should be added to the regular suite of contaminants analyzed

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Human Use Pharmaceuticals

- "Extremely little is known about the effects of these substances on non-target organisms, many of which have different metabolic pathways and different potential receptors.
- By their nature, pharmaceuticals are designed to be highly bioactive -- many exquisitely so.
- What little that is known serves to show that rather low concentrations at least have the potential to exert substantive effects on aquatic life."

Christian Daughton, Ph.D.
U.S. EPA, Office of Research & Development

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Human-Use Pharmaceuticals

- Estimated 3.84 billion prescriptions written in 2008 for pharmaceuticals in the U.S. (IMS Health, 2009)
- 1.4 million kg of antimicrobials used in human medicine in the U.S. every year (Mellon et al., 2001)
- Development of antibiotic-resistant pathogenic bacteria in the environment is a concern
- Pharmaceuticals continuously released into the environment
- Potential to produce the same exposure as truly persistent compounds

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Human-Use Pharmaceuticals

- Ingestion and excretion thought to be the main pathway for introduction to the environment
- Incomplete removal by wastewater treatment plants (WWTPs) can result in the discharge of pharmaceuticals to surface waters
- Leachate into surface or ground waters from septic systems is also a likely source
- Biologically active compounds
- Most work done to date to assess the aquatic occurrence of pharmaceuticals has been in freshwater systems, less work in estuarine and marine waters

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2002 Pilot Project

- Assess the presence of a group of 24 pharmaceuticals and related compounds in the Chesapeake Bay
- Samples taken in conjunction with a NOAA project on sediment toxicity
- Water samples extracted at the University of Maryland
  - Caveat - sample extracts stored (frozen) for approximately 6 months prior to analysis
  - Conservative estimates of the original concentrations of pharmaceuticals in the water samples
- Extracts analyzed by the USGS, National Water Quality Laboratory in Denver, CO

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Sampling Strategy

- Fourteen sites targeted in the Chesapeake Bay in 2002
- Water samples collected adjacent to, and in some cases downstream of, WWTPs in order to have the best chance of detection
- Composite (surface and near bottom) water samples

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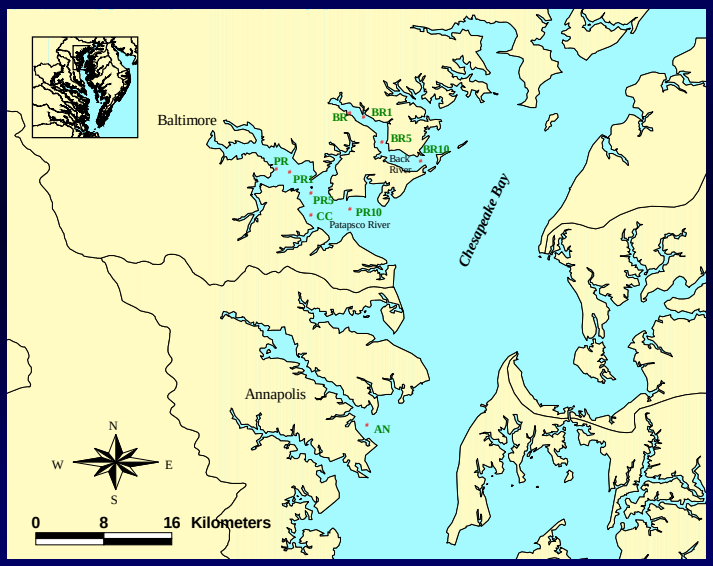
Sampling Sites – Northern Chesapeake Bay



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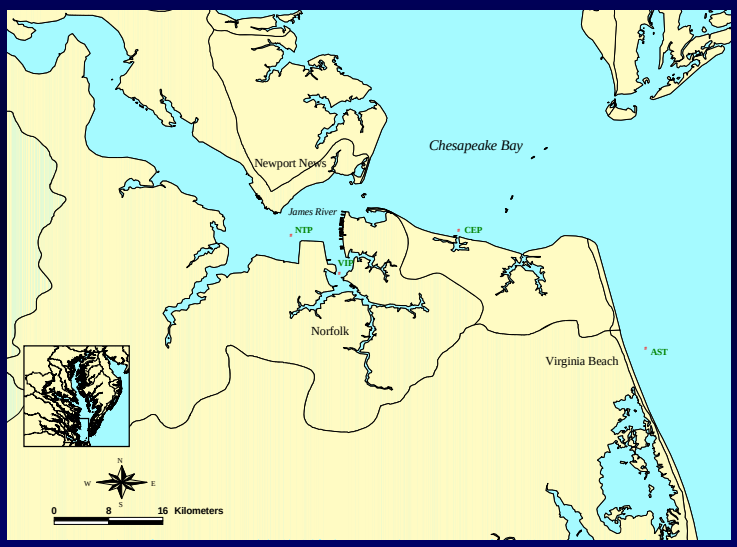
Sampling Sites – Southern Chesapeake Bay



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Field Work

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Extraction and Analysis

Solid phase extraction

HPLC/ESI-MS

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Pharmaceuticals Analyzed

Compound	Use	Brand/Common Name	MW	CAS
1,7-dimethylxanthine	Caffeine metabolite	Paraxanthine	180.16	611-59-6
Acetaminophen	Analgesic and antipyretic	Tylenol®	151.17	103-90-2
Azithromycin	Antibiotic	Zithromax®	748.88	83905-01-5
Caffeine	Stimulant	Caffeine	194.19	58-08-2
Carbamazepine	Antiepileptic, antidepressant	Tegretol®	236.27	298-46-4
Cimetidine	Antacid	Pepcid®	252.34	51481-61-9
Codeine	Analgesic	Codeine	299.36	76-57-3
Cotinine	Nicotine metabolite	Cotinine	176.22	486-56-6
Dehydronifedipine	Antianginal	Procardia® metabolite	344.32	67035-22-7
Diltiazem	Antianginal	Cardizem®	450.98	33286-22-5
Diphenhydramine	Antihistamine	Benadryl®	291.82	147-24-0
Erythromycin	Antibiotic	E-mycin®	733.93	114-07-8

Abbreviations: CAS, Chemical Abstract Service; MW, molecular weight; ®, Registered trademark

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Pharmaceuticals Analyzed

Compound	Use	Brand/Common Name	MW	CAS
Fluoxetine	Antidepressant	Prozac®	345.8	54910-89-3
Gemfibrozil	Lipid regulator	Lopid®	250.35	25812-30-0
Ibuprofen	Analgesic and antipyretic	Motrin®	351.83	15687-27-1
Metformin	Antidiabetic	Glucophage®	129.17	657-24-9
Miconazole	Antifungal	Micatin®	416.12	22916-47-8
Paroxetine metabolite	Antidepressant	Paxil® metabolite	-	-
Ranitidine	Antacid	Zantac®	350.87	66357-35-5
Salbutamol	Antiasthmatic	Proventil®	239.3	51022-70-9
Sulfamethoxazole	Antibiotic	Bactrim®	253.28	723-46-6
Thiabendazole	Anthelmintic	Mintezol®	201.26	148-79-8
Trimethoprim	Antibiotic	Proloprim®	290.3	738-70-5
Warfarin	Anticoagulant	Coumadin®	308.33	129-06-6

Abbreviations: CAS, Chemical Abstract Service; MW, molecular weight; ®, Registered trademark

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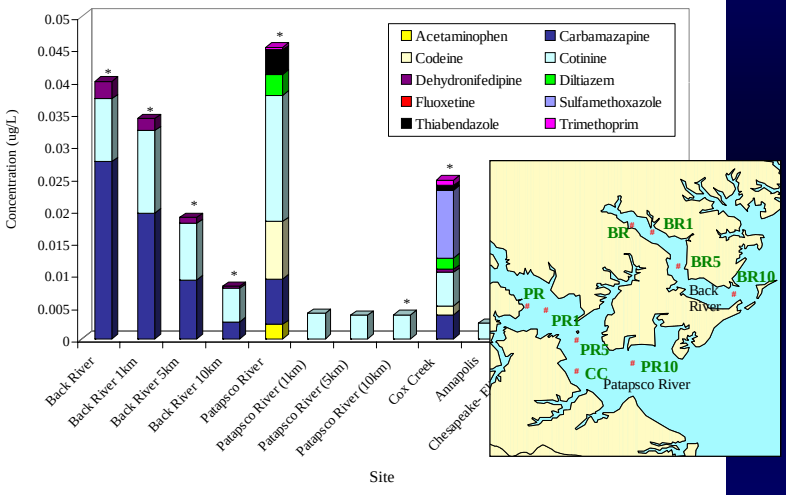
Summary of Results



*Erythromycin-H_2O, detected but not quantified

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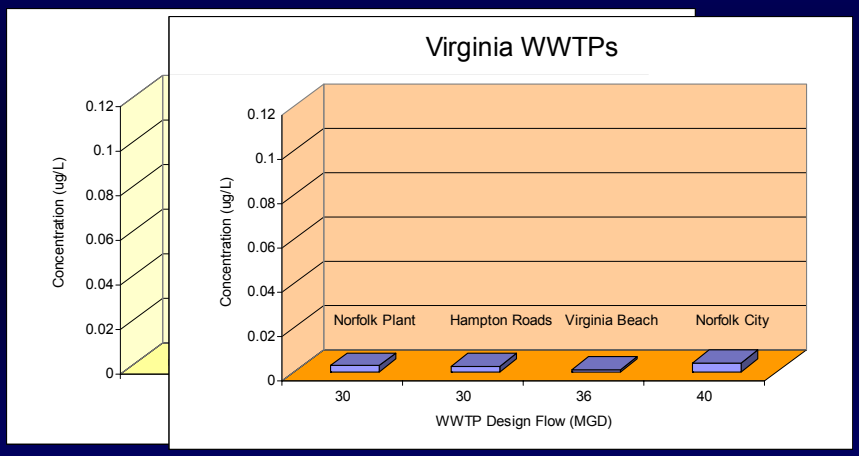
Comparison of Maryland and Virginia WWTP sites



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Conclusions

- A number of frequently prescribed human-use pharmaceuticals were detectable in water samples collected adjacent to WWTPs in the Chesapeake Bay
- Three compounds were 10 km downstream of the Back River WWTP
- Concentrations were in the sub µg/L range
- Effects (if any) on aquatic organisms within the environment is unknown
- An important first step is to document which pharmaceuticals are present and at what concentrations

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- Steve Werner of the National Water Quality Laboratory, USGS Denver
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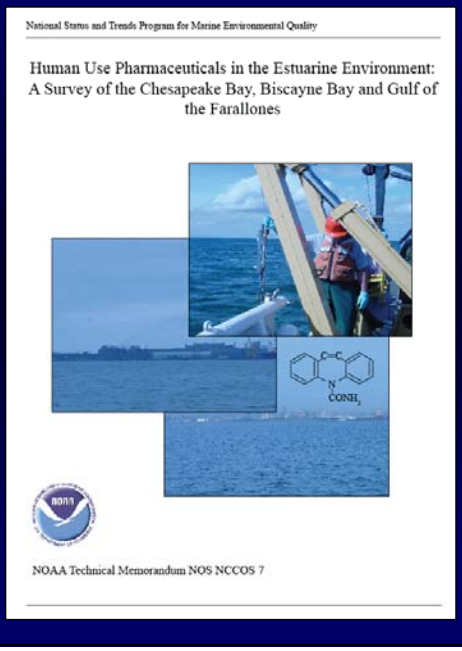
Report available

(also includes Biscayne Bay and Gulf of the Farallones)

http://www.ccma.nos.noaa.gov/publications/HumanUsePharma.pdf


National Status and Trends Program for Marine Environmental Quality

Human Use Pharmaceuticals in the Estuarine Environment: A Survey of the Chesapeake Bay, Biscayne Bay and Gulf of the Farallones

NOAA Technical Memorandum NOS NCCOS 7