



# ***Peering into the Shadows of Chemical Space. Emerging Contaminants and Environmental Science: Is either being served by the other?***

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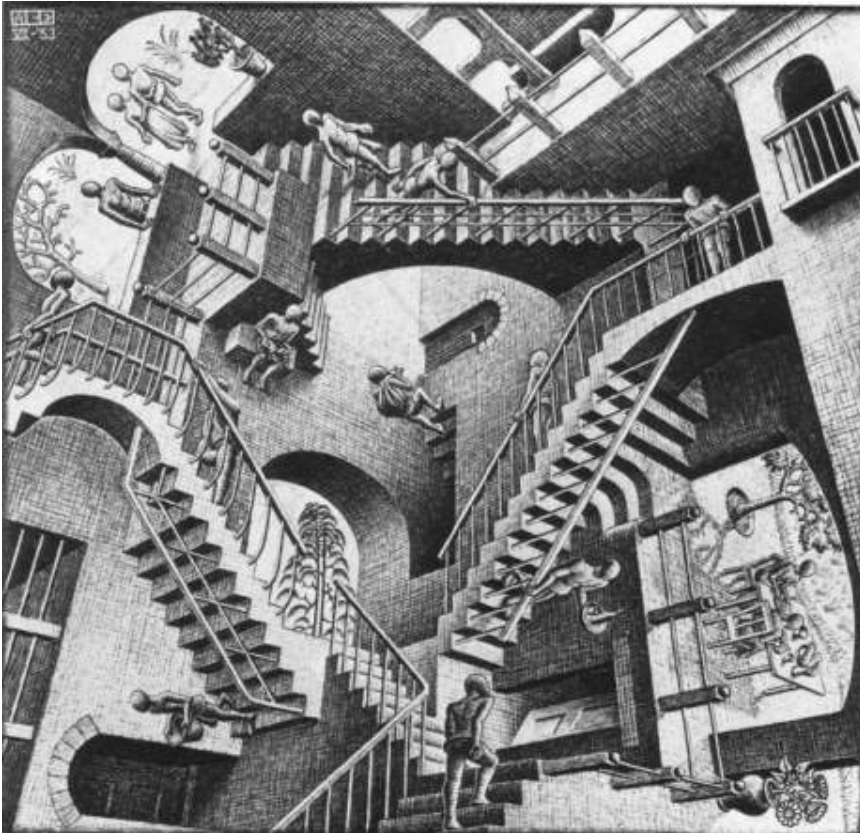
**Office of Research and Development**

National Exposure Research Laboratory, Environmental Sciences Division, Las Vegas, Nevada

**Does anyone know what an  
*Emerging Contaminant* really is?**



# *Emerging Contaminants ...*



*... depend on  
one's perspective*

# What Makes a “Contaminant of Emerging Concern”?

ECs include more than simply chemicals previously not known to occur in the environment.

ECs also include chemicals already known to occur but now displaying new characteristics not previously suspected or recognized, such as those involving:

- origin or source
- location (e.g., “out-of-place” chemicals; “chemical weeds”)
- concentrations or levels
- transformation and fate pathways
- exposure routes
- effects pathways or endpoints



## *Slicing the Pollutant Pie*

Environmental contaminants can be categorized across many dimensions.



A host of acronyms describe different slices, often breeding confusion.

# *Acronym Anarchy*

EDCs – CMRs – PBTs – vPvB – POPs  
– OWCs – PPCPs – PhACs – EPOCs  
– COCs – COPCs – CECs – XOCs –  
HPVs – PDPs – HDPs – POHOs –  
SVHCs

ppm – ppb – ppt – ppq  
micro – nano – pico

## The Confusing Terminology of Chemical Pollutant Classes

| <b>Class or Grouping</b>   | <b>Grouped According to:</b>  |
|--|---|
| <b>EDC</b> (Endocrine Disrupting Chemical)<br><b>CMR</b> (Carcinogenic, Mutagenic, toxic to Reproduction)  | toxicological mode of action or endpoint  |
| <b>PBT</b> (Persistent, Bioaccumulative Toxic)<br><b>vPvB</b> (very Persistent, very Bioaccumulative)<br><b>POP</b> (Persistent Organic Pollutant)   | environmental properties  |
| <b>micro-pollutants; micro-constituents</b><br><b>trace pollutants/residues</b>  | frequency/level of occurrence   |
| <b>OWCs</b> (Organic Wastewater Contaminants)  | location of occurrence  |
| <b>PPCPs</b> (also PhACs, which is a subset)   | type of intended usage  |
| <b>priority pollutants</b> and others  | regulation  |
| <b>unregulated pollutants</b>  | absence of regulation   |
| <b>“chemical weeds”</b>  | public perception ("out-of-place chemicals")  |
| <b>emerging contaminants/pollutants</b><br><b>EPOCs</b> (emerging pollutants of concern)<br><b>COCs</b> (chemicals of concern)<br><b>COPCs</b> (chemicals of potential concern)<br><b>CECs</b> (chemicals of emerging concern) | novelty, fad, timeliness, or new concern  |
| <b>xenobiotics, exotics</b><br><b>xenobiotic organic compounds (XOCs)</b>  | foreign versus endogenous   |
| <b>toxicants, toxins, toxics, perturbogens</b> (agonists, antagonists, activators, repressors, inhibitors, regulators, modulators)   | overall toxicity ( <i>note</i> : "toxins" comprise a special subset of toxicants - those naturally synthesized, primarily proteins; "toxics" is jargon for "toxicants") |
| <b>HPV</b> (high production volume) chemicals  | quantity (manufactured/imported in US $\geq 1$ million pounds/year)   |
| <b>PDPs</b> or <b>HDPs</b> (population- or human-derived pollutants/constituents)<br><b>POHO</b> (pollutants of human origin)  | source or origin  |

Adapted and extended by Daughton from: *Renewable Resources Journal*, 2005, 23(4):6-23.

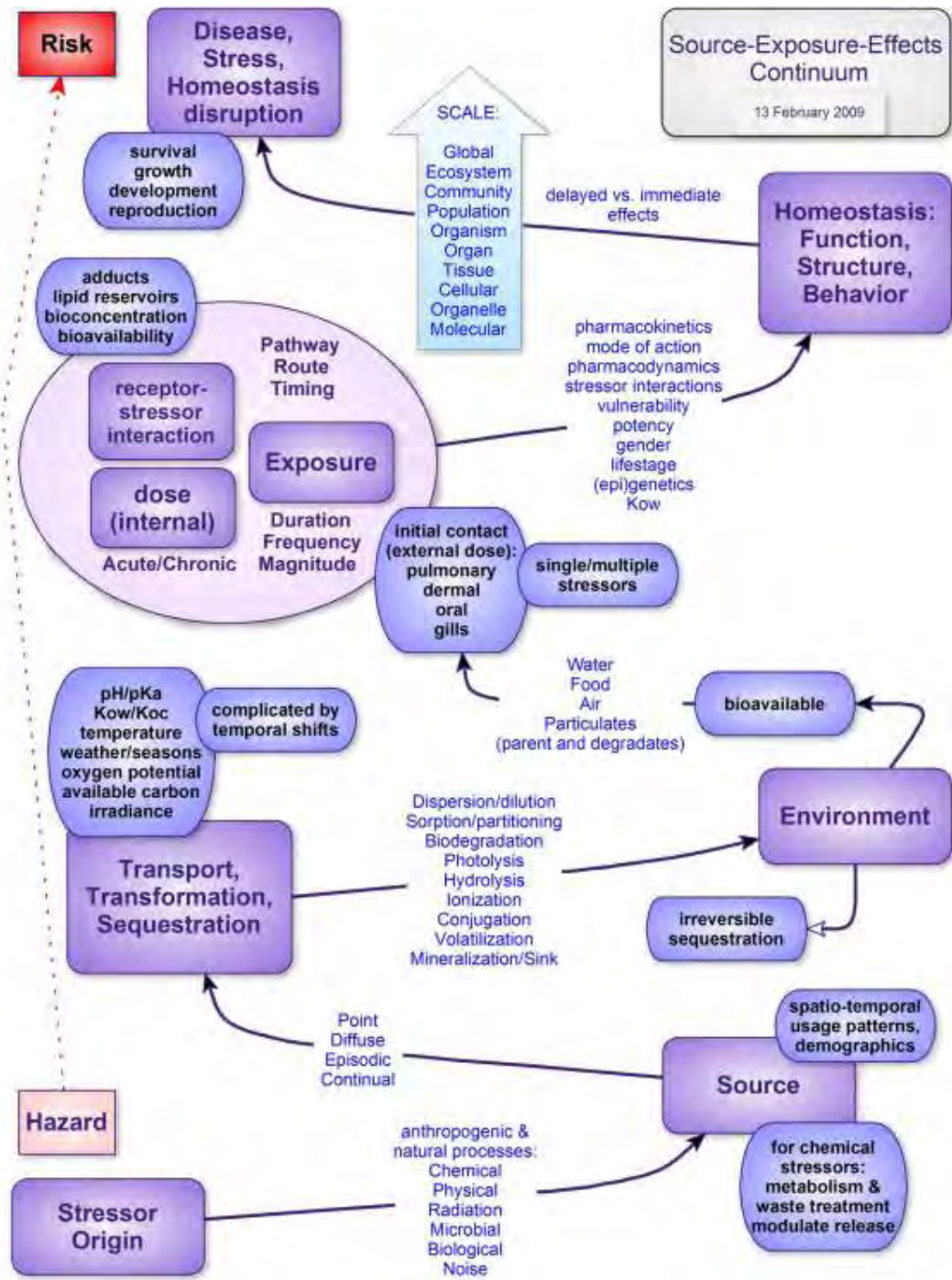
# Context of “*Emerging*”

The term "emerging" can be applied to any part of the continuum that spans from stressors to exposure and effects. It applies not just to chemicals, but also to any previously unrecognized, unexpected, or under-appreciated aspect of environmental science.

The study of "emerging" contaminants is really one of more carefully examining the interface between humans and the environment; and of continually expanding the conventional boundaries surrounding all facets of the source-to-effects continuum.

The term "ECs" causes confusion because it pertains not just to chemicals themselves but also to the many dimensions associated with their properties and the source-to-effects continuum.





## *Some EPA Perspective*

Statement of **Peter S. Silva** (confirmed, June 2009, as Assistant Administrator for the Office of Water) at hearing of Nominations Committee on Environment and Public Works United States Senate:

"... despite the considerable progress we have made in the last three decades, we now see additional challenges have arisen in the areas of non-point source pollution and in new emerging pollutants of concern." (12 May 2009)

"Managing Chemical Risks" is one of EPA's five new priority areas identified by EPA's new Administrator, **Lisa Jackson**:

"It is clear that we are not doing an adequate job of assessing and managing the risks of chemicals in consumer products, the workplace and the environment." (23 January 2009)

## *Some EPA Perspective*

**Peter S. Silva** (American Water Works Association, Annual Conference and Exposition, 15 June 2009, San Diego)

"There is a growing nexus between the way we supply water and how we face environmental impacts."

"Even direct potable reuse is being considered."



# "Emerging" Contaminants

Most "emerging" contaminants are not new to the environment

- Two major sources for contaminants that are truly "new" to the environment:
  - Chemicals newly introduced to commerce (e.g., new approved or unapproved drugs, pesticides, nanomaterial).
  - By-products of new anthropogenic processes (e.g., nanotechnology).
- Previously unrecognized pollutants can come to our attention as a result of:
  - New advances in chemical analysis (e.g., "non-target" identification).
  - Ability to detect existing pollutants at ever-lower concentrations (single-molecule detection is now possible).
  - Exploring exposure sources not previously examined (e.g., certain foods as a significant source of acrylamide; estrogens in plastics; PPCPs in biosolids; alkaloids in pyrolysis products from controlled forest burns).



# examples

Old pollutants in new places: organohalogenes in dolphin & seal brain; conventional POPs and their metabolites in tissues never before explored.

Old chemicals as new suspects: tungsten: monotungstate, polytungstate, and heteropolytungstates

Toxicants masquerading as "inert" ingredients: polyethoxylated tallow amine used as adjuvant for formulating glyphosate; shows high aquatic toxicity and toxicity in human cell cultures.

Occurring at levels or locations never before observed: APIs in drug manufacturing waste streams. Locations indicative of possible acts of sabotage.

New pollutants: perfluorinated phosphonic acids (in waters); unapproved drugs

Source or origin: marine production of halogenated organics;

alkaloids in particulates/aerosols from controlled burns in forests (pyrolytic conditions);

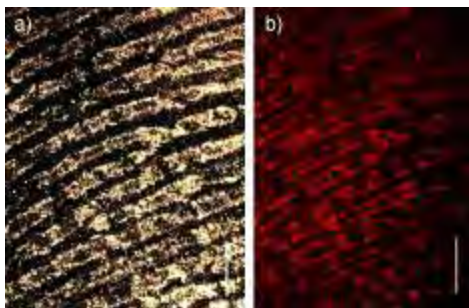
melamine in infant formula, originating as metabolite of cyromazine, an insecticide used on crops and forage

Route of transport to the environment (contributing to exposure risk): excretion of drugs via the skin (sweat); illicit drugs in ambient air

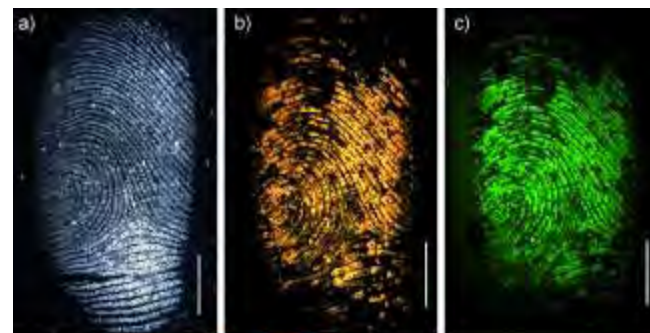
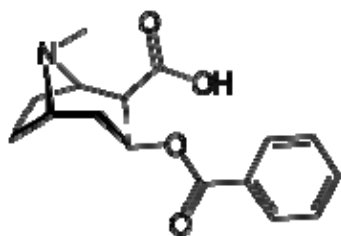
Exposure pathway: olfactory nerve of fish



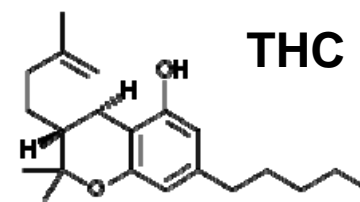
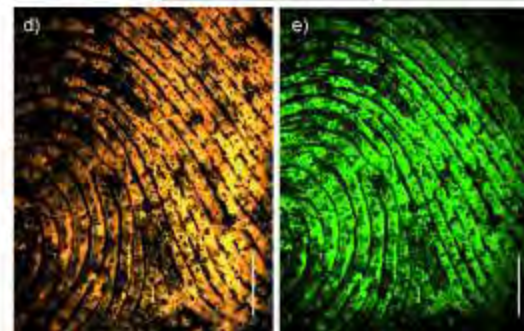
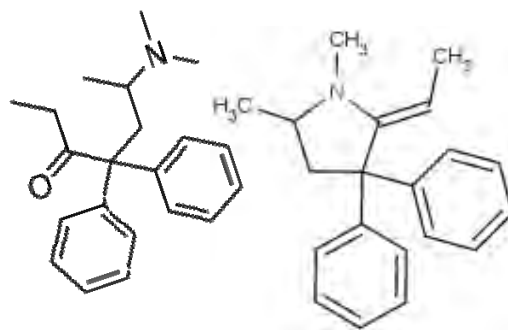
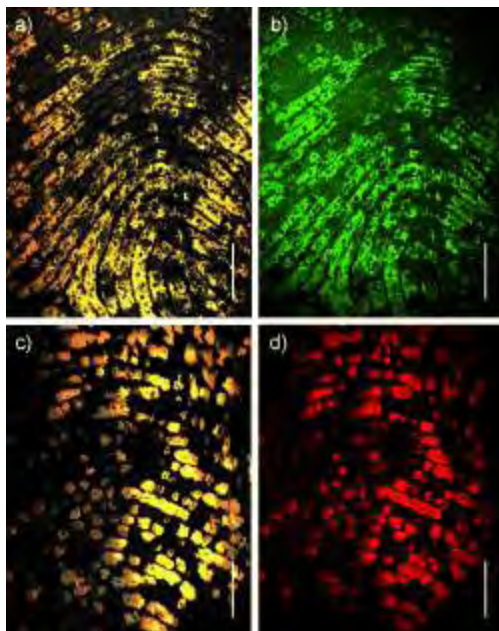
# Fingerprints as contributors of environmental contaminants



benzoylecgonine



methadone & EDDP



*benzoylecgonine* (major cocaine metabolite)

*EDDP*: 2-ethylidene-1,5-dimethyl-3,3-diphenylpyrrolidine (major methadone metabolite)

*THC*:  $\Delta^9$ -tetrahydrocannabinol (main psychoactive of marijuana)

Hazarika P, Jickells SM, Wolff K, Russell DA (2008) Imaging of Latent Fingerprints through the Detection of Drugs and Metabolites. *Angew Chem Int Ed* 47:10167-10170.

*Challenges for environmental chemical analysis:  
Complexities in identifying and monitoring chemical unknowns*

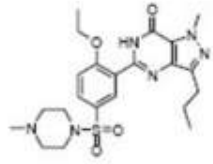
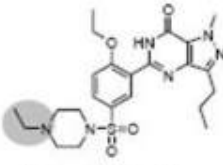
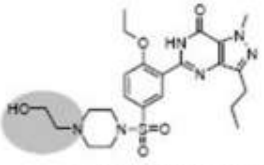
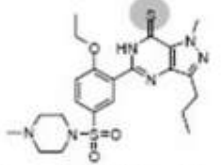
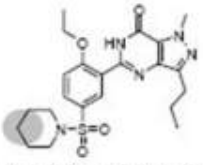
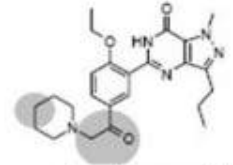
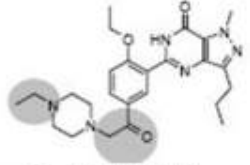
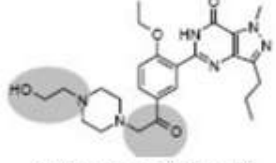
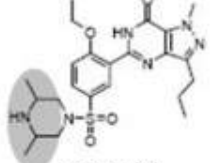
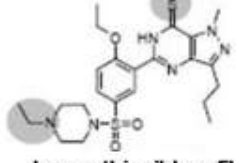
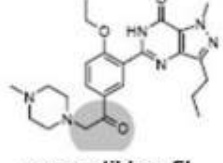
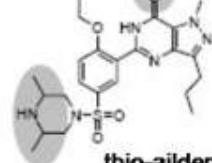
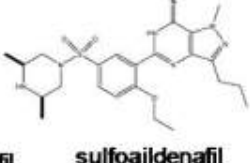
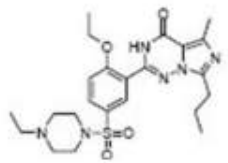
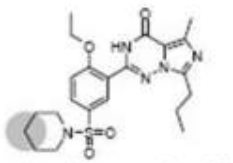
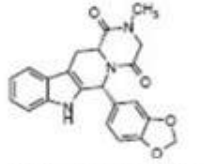
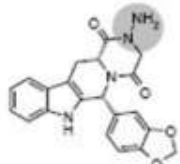


- **Chiral enantiomers**: long proven important for certain pharmaceuticals and pesticides (e.g., improved profiles for effectiveness and toxicity versus hydrogen analogs). Subject to enantiomeric enrichment via microbial degradation.
- **Nanomaterials**: pose enormous challenges for chemical characterization because of diversity of structures and possible complexity of weathered products.
- **Unapproved drugs**: unapproved analogs of registered drugs; sold OTC in adulterated herbal supplements and in fraudulent prescription medications.

# Designer Drugs: Environmental Contaminant Unknowns

Undeclared drug active ingredients (new molecules) – known or unknown to FDA.  
Adulterants in herbal supplements or OTC/Rx drugs, often at high levels.

*Example:* analogs of the approved phosphodiesterase type 5 (PDE-5) inhibitors (used primarily in the treatment of erectile dysfunction), such as sildenafil, vardenafil, and tadalafil.

| Registered APIs  | Analogues identified  |  |   |
|--|---|--|---|
| <br><b>sildenafil (Viagra®)</b>    | <br><b>homosildenafil</b>          | <br><b>hydroxyhomosildenafil</b>    | <br><b>thiosildenafil</b>      |
|  | <br><b>piperidino-sildenafil</b>   | <br><b>piperidino-acetildenafil</b> | <br><b>acetildenafil</b>       |
|  | <br><b>hydroxy-acetildenafil</b>   | <br><b>aildenafil</b>               | <br><b>homo-thiosildenafil</b> |
|  | <br><b>nor-acetildenafil</b>       | <br><b>thio-aildenafil</b>          | <br><b>sulfoaildenafil</b>     |
| <br><b>vardenafil (Levitra®)</b> | <br><b>piperidino-vardenafil</b> |  |   |
| <br><b>tadalafil (Cialis®)</b>   | <br><b>amino-tadalafil</b>       |  |   |

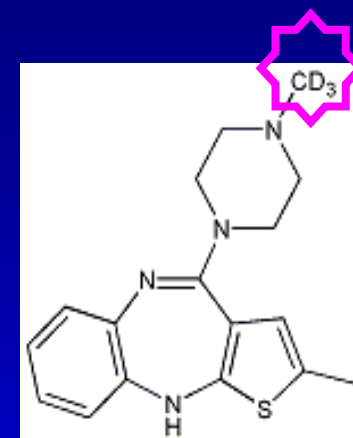
Herbal supplements:  
*Y-4ever*  
*Powermania*

Herbal supplements:  
*Zencore Tabs*  
*LibieXtreme*  
*Libimax X*

B.J. Venhuis, D.M. Barends, M.E. Zwaagstra, and D. de Kaste "Recent developments in counterfeits and imitations of Viagra, Cialis and Levitra: A 2005-2006 update," 2007, RIVM (Netherlands National Institute for Public Health and the Environment), RIVM Report 370030001/2007, Bilthoven, the Netherlands, 61 pp; <http://www.rivm.nl/bibliotheek/rapporten/370030001.pdf>

## Challenges for environmental chemical analysis: Complexities in identifying and monitoring chemical unknowns

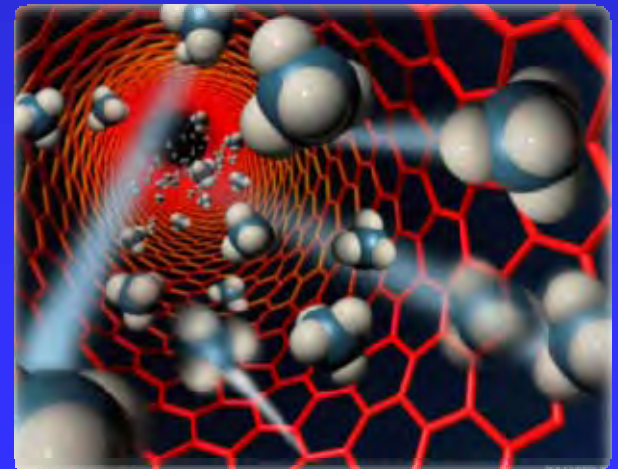
- **Isotopic substitution**: *deuterated analogs* of pharmaceuticals and pesticides long known for altered pharmacokinetics/pharmacodynamics because of kinetic isotope effect.
  - Pharmacologic deuteration yields drugs with greater stability. Can facilitate longer half-lives (increased duration of action from hindered first-pass metabolism); fewer side effects from lower dose, "metabolic switching," and reduced drug-drug interactions.
  - Major unknown for deuterated drugs: alteration of environmental fate & transport?
  - Major new analytical challenge: each API can have one or more deuterated analogs, increasing the numbers of potential analytes. But moreover, these analogs would greatly hinder the identification of each other if they cannot be physically separated prior to mass spectral detection (a result of radically altered isotopic abundances).



# ***Structurally Undefinable Xenobiotics***

## **Importance of Developing the Means to Evaluate and Monitor "Undefinable" Xenobiotics**

- The widespread emergence in consumer products of new **nanomaterials** poses a host of currently unanswerable questions regarding their environmental chemistry and environmental toxicology.
- In particular, the natural weathering or environmental transformation of these materials holds the potential to yield numerous additional, **structurally undefinable products**.
- **Nanomaterials pose numerous, currently intractable challenges to their monitoring in the environment.**



# It's Been Only 10 Years

Perhaps the first published documents to prominently feature the term *emerging contaminants* were the 1999 report from the 1998 NRC workshop ("Identifying Future Drinking Water Contaminants") and several 1999 reports by USGS.



*1st North American Conference (ACS) on  
Pharmaceuticals and Personal Care Products  
in the Environment -- San Francisco, 27 March 2000*



# *Environmental Contaminants*

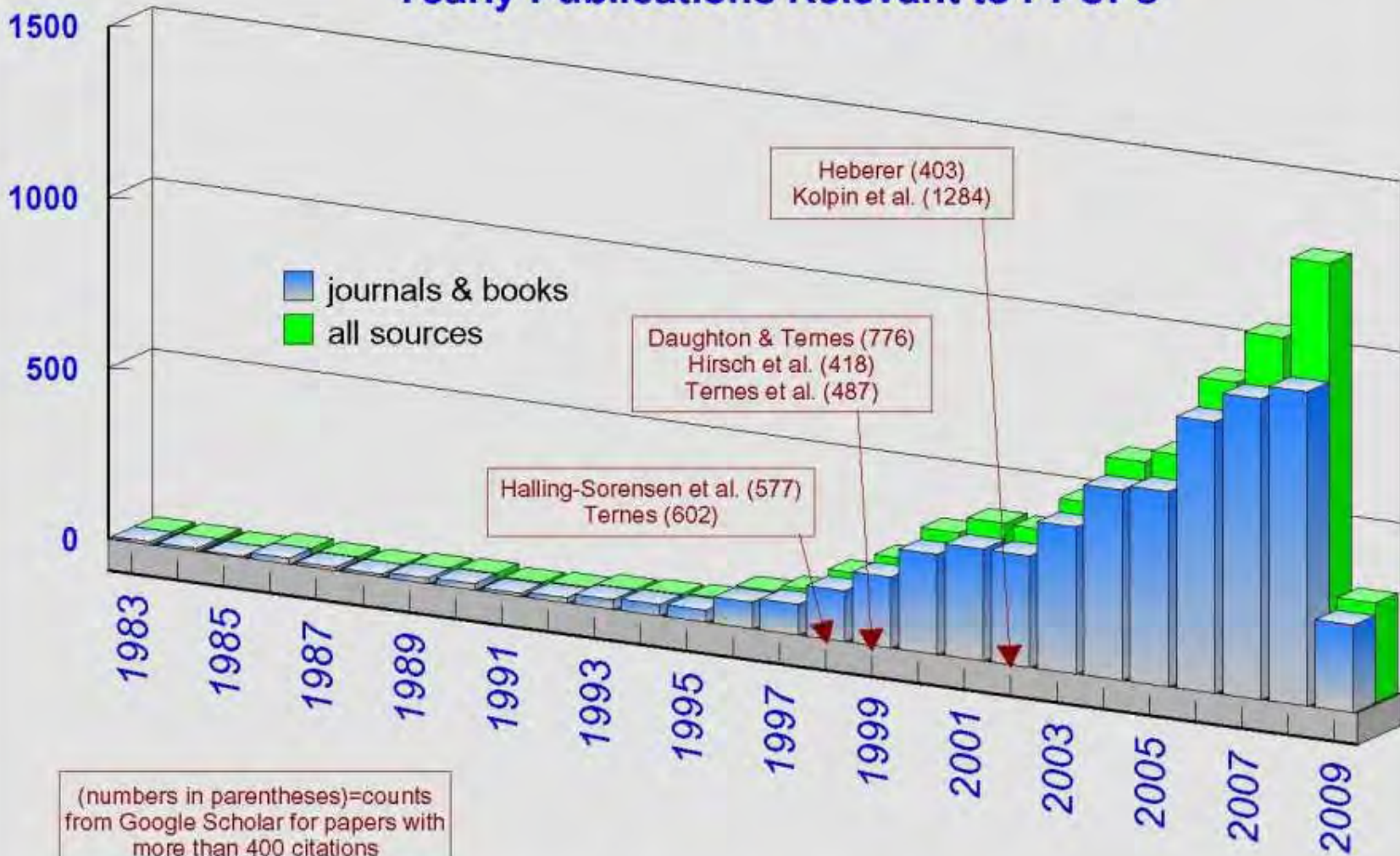
## - Status Check -



Passage of a decade marks an excellent time to reflect on a number of questions surrounding this rather ill-defined but broadly used term.

- Has the term served us well?
- Do we share a common understanding as to what an "emerging" contaminant might be?
- Is it leading us where we need to be in terms of a scientific discipline?
- What message does it convey to the public, legislators, and regulators?
- Are we perhaps suffering from its overuse – having become an obligatory adjective?
- What about the galaxies of other chemicals that currently evade detection and cannot yet be classified as “emerging”?

## Yearly Publications Relevant to PPCPs



note: data for 2009 only through first 8 weeks

# A More Accurate Birth Date – 150 years ago

George P. Marsh: forerunner to the modern environmental movement. Over 150 years ago, recognized that interconnections pervade all of nature.

Birth of the idea of incremental, cumulative impacts from small forces. Foretold the new paradigm of countless point-sources combining to serve as a unified dispersed source: **the significance of the combined but miniscule actions, activities, and behaviors of multitudes of individuals.**

Perhaps the first scientific examination and documentation of the thesis that harm accumulates, regardless of the seeming insignificance of each act of harm.



Marsh GP. 1864. *Man and nature; or, Physical geography as modified by human action.*  
C. Scribner, New York.

## 1864: Birth of the idea of incremental, cumulative impacts [or - Small Can Be as Big as Big]

From the chapter:

**"Nothing Small in Nature."**

Marsh GP. 1864.

Man and nature; or, Physical geography as modified by human action.

C. Scribner, New York.



“No atom can be disturbed in place, or undergo any change of temperature, of electrical state, or other material condition, without affecting, by attraction or repulsion or other communication, the surrounding atoms. These, again, by the same law, transmit the influence to other atoms, and the impulse thus given extends through the whole material universe.” “...reaction does not restore disturbed atoms to their former place and condition, and consequently the effects of the least material change are never cancelled, but in some way perpetuated, so that *no action can take place in ... nature, without leaving all matter in a different state from what it would have been if such action had not occurred.*”

## *Emerging Challenges from Emerging Contaminants*

- Increasingly advanced methods of analysis allow peering into the shadows of chemical space with ever-greater magnification and clarity.
- At ever-lower exposure levels, it may no longer be possible to deconvolute a purported effect from its incidence as ambient (natural) background.
- Ever-lower detection limits pose increasingly greater challenges for assessing, communicating, and ameliorating ever-diminishing risks.
- Where do these risks reside within the overall scope of larger environmental concerns?



# Complex Mixtures of Chemical Stressors at Low Concentrations:

*Pose Major Challenges for Measurement and for  
Assessing Exposure and Biological Effects*



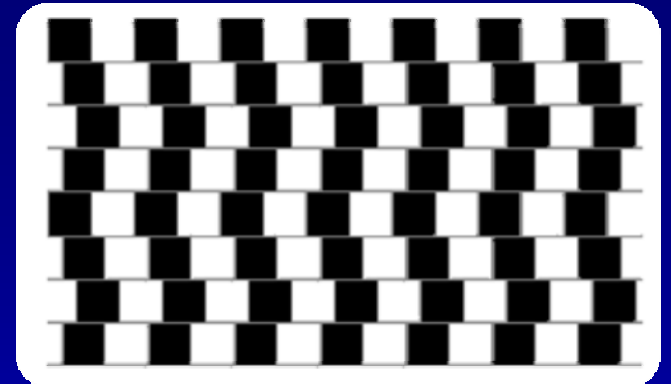
***What portion of overall risk is  
contributed by unregulated  
contaminants?***



## *Public Perception*

Perceive only two reasons that ECs occur in the environment:

1. Newly introduced to commerce
  2. We failed to detect them up until now
- ECs serve to highlight the fact that our environmental monitoring system is reactive rather than anticipatory.
  - The term "ECs" tends to alarm the public, possibly cultivating a negative image of failure on the part of environmental scientists. Perception does not align with reality.
  - After all, the reason that a pollutant becomes known as "emerging" is simply because of our failure to anticipate or predict. In a perfect world, we should be able to identify and prevent the "emergence" of chemicals.
  - Problem with continual identification of yet more ECs with no actions to prevent future ones.



# The Chemical Universe

We live in a chemical sea of continually changing composition – comprising both anthropogenic and naturally occurring chemical stressors.

Unlike biota, chemical pollutants have no boundaries in their global distribution –  
“*everything is everywhere,*” only the concentrations vary.



# The Chemical Universe

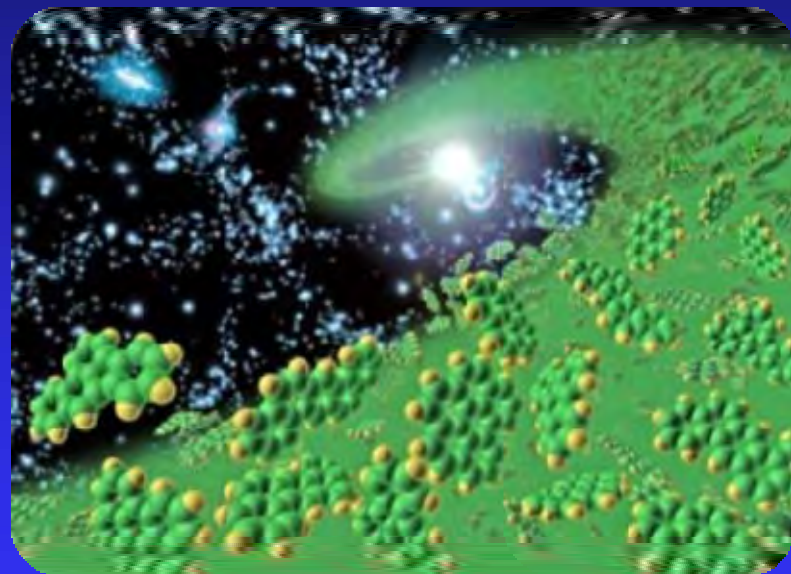
## The *KNOWN* Universe

As of July 2009, over 48 million organic and inorganic substances had been assigned CAS RNs.

(indexed by the American Chemical Society's Chemical Abstracts Service in their CAS Registry; excluding bio-sequences such as proteins and nucleotides:

<http://www.cas.org/expertise/cascontent/registry/regsys.html>)

- Of these millions of known chemicals, nearly 35 million were commercially available.
- Of these, fewer than a quarter million (249,000) are inventoried or regulated by numerous government bodies worldwide - - representing less than 0.8% of those that are commercially available or less than 0.6% of the known universe of chemicals.
- Approximately 12,000 new substances are added each day.



# The Chemical Universe

## The *POTENTIAL* Universe

While the *KNOWN* universe of commercial chemicals might seem large (35 million), the universe of *POTENTIAL* chemicals (those that could possibly be synthesized and those that already exist but which have not yet been identified) is unimaginably large.

How many distinct organic chemical entities could hypothetically be synthesized and added to a seemingly limitless, ever-expanding chemical universe?

By limiting synthesis strictly to combinations of 30 atoms of just C, N, O, or S, **more than  $10^{60}$  structures are possible !**

Expanding the allowable elements to other heteroatoms (e.g., P and halogens) or deuterium, **the limits to the numbers of possible structures in “chemical space” defies imagination.**

# The Chemical Universe

The largest virtual chemical database yet reported comprises small drug-like molecules - - a total of over 977,000,000 structures (Blum and Reymond, 2009).

Restricted to organic molecules containing fewer than 14 atoms of C, N, O, and S (and limited types of Cl-substituted molecules). Excluded likely substituents such as F, Br, I, P, Si, metals, and most Cl.

Database represents the enormously large numbers of chemicals that could possibly be synthesized just from a very limited spectrum of types of elements and numbers of atoms.

Blum, L. C., and J.-L. Reymond. 2009. 970 Million Druglike Small Molecules for Virtual Screening in the Chemical Universe Database GDB-13. *J. Am. Chem. Soc.* 131 (25):8732-8733.

# 200 Top-Selling Prescribed Drugs (2006)

|     |     |     |     |     |     |     |     |     |     |     |     |     |     |     |     |     |     |     |     |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| 1   | 2   | 3   | 4   | 5   | 6   | 7   | 8   | 9   | 10  | 11  | 12  | 13  | 14  | 15  | 16  | 17  | 18  | 19  | 20  |
| 21  | 22  | 23  | 24  | 25  | 26  | 27  | 28  | 29  | 30  | 31  | 32  | 33  | 34  | 35  | 36  | 37  | 38  | 39  | 40  |
| 41  | 42  | 43  | 44  | 45  | 46  | 47  | 48  | 49  | 50  | 51  | 52  | 53  | 54  | 55  | 56  | 57  | 58  | 59  | 60  |
| 61  | 62  | 63  | 64  | 65  | 66  | 67  | 68  | 69  | 70  | 71  | 72  | 73  | 74  | 75  | 76  | 77  | 78  | 79  | 80  |
| 81  | 82  | 83  | 84  | 85  | 86  | 87  | 88  | 89  | 90  | 91  | 92  | 93  | 94  | 95  | 96  | 97  | 98  | 99  | 100 |
| 101 | 102 | 103 | 104 | 105 | 106 | 107 | 108 | 109 | 110 | 111 | 112 | 113 | 114 | 115 | 116 | 117 | 118 | 119 | 120 |
| 121 | 122 | 123 | 124 | 125 | 126 | 127 | 128 | 129 | 130 | 131 | 132 | 133 | 134 | 135 | 136 | 137 | 138 | 139 | 140 |
| 141 | 142 | 143 | 144 | 145 | 146 | 147 | 148 | 149 | 150 | 151 | 152 | 153 | 154 | 155 | 156 | 157 | 158 | 159 | 160 |
| 161 | 162 | 163 | 164 | 165 | 166 | 167 | 168 | 169 | 170 | 171 | 172 | 173 | 174 | 175 | 176 | 177 | 178 | 179 | 180 |
| 181 | 182 | 183 | 184 | 185 | 186 | 187 | 188 | 189 | 190 | 191 | 192 | 193 | 194 | 195 | 196 | 197 | 198 | 199 | 200 |

# Black Holes in Exploring the Universe Chemical by Chemical

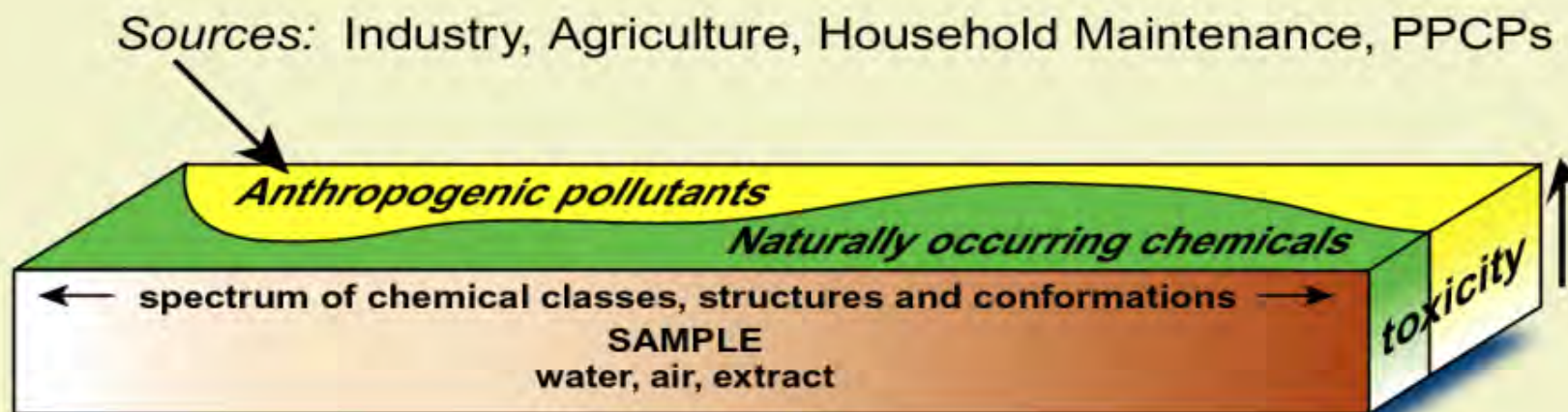


- Traditional chemical-by-chemical approach for evaluating risk of new and existing chemicals is not sustainable (speed, complexity, cost)
- Extensive environmental and toxicological data exist for very few chemicals
- New paradigm will be required to cope with the backlog of existing chemicals and the sheer number of possible new chemicals
- Realization of the critical importance of multi-generational, simultaneous exposure to individual trace levels of multitudes of chemical stressors
- Subtle, delayed-onset effects
- Chemicals cannot be prioritized based solely on commercial volume and conventional toxicity testing
- Assessment of hazard (both virtual and empirical) will require extremely high throughput



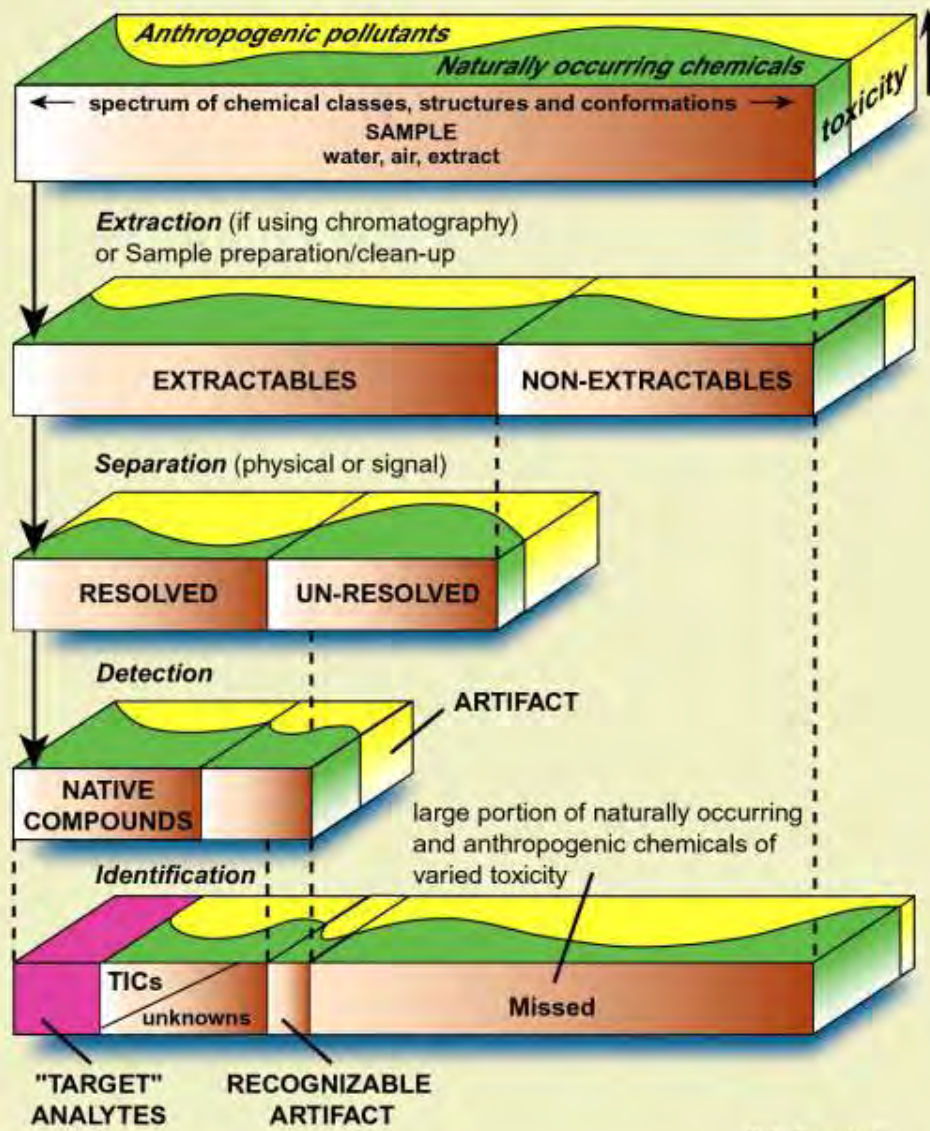
**Lessons learned from emerging contaminants will undoubtedly play a major role in the future of risk assessment.**

# Universe of Chemicals in the Environment



From: Daughton, C.G. "Cradle-to-Cradle Stewardship of Drugs for Minimizing Their Environmental Disposition while Promoting Human Health. I. Rationale for and Avenues toward a Green Pharmacy," *Environ. Health Perspect.* **2003**, 111(5):757-774; available: <http://www.epa.gov/nerlesd1/bios/daughton/green1.pdf>.

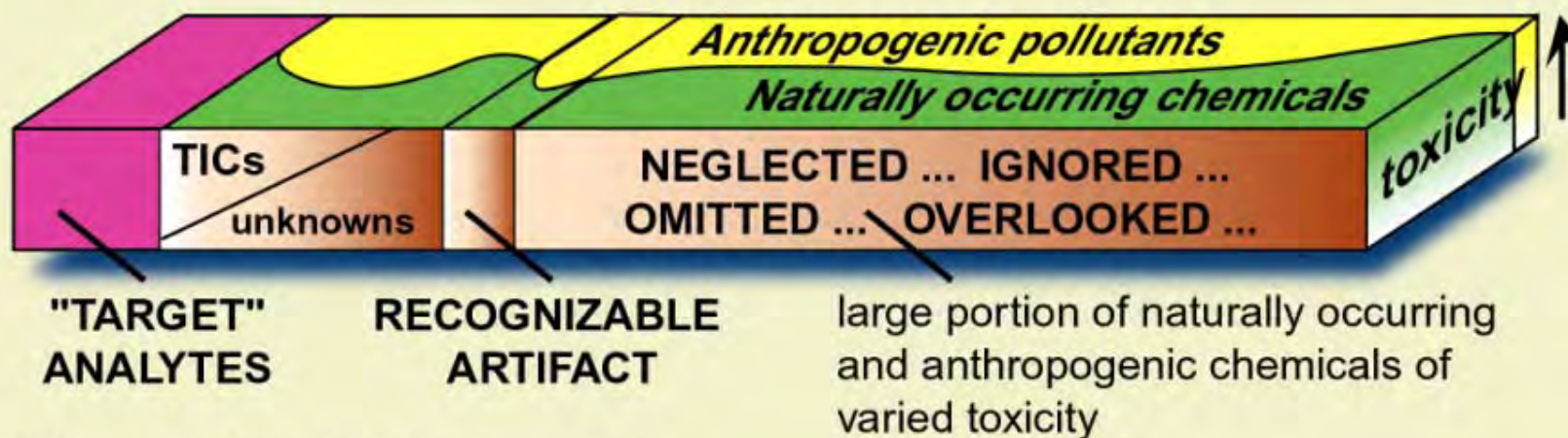
# Limitations and Complexity of Environmental Chemical Analysis



TICs = tentatively identified compounds

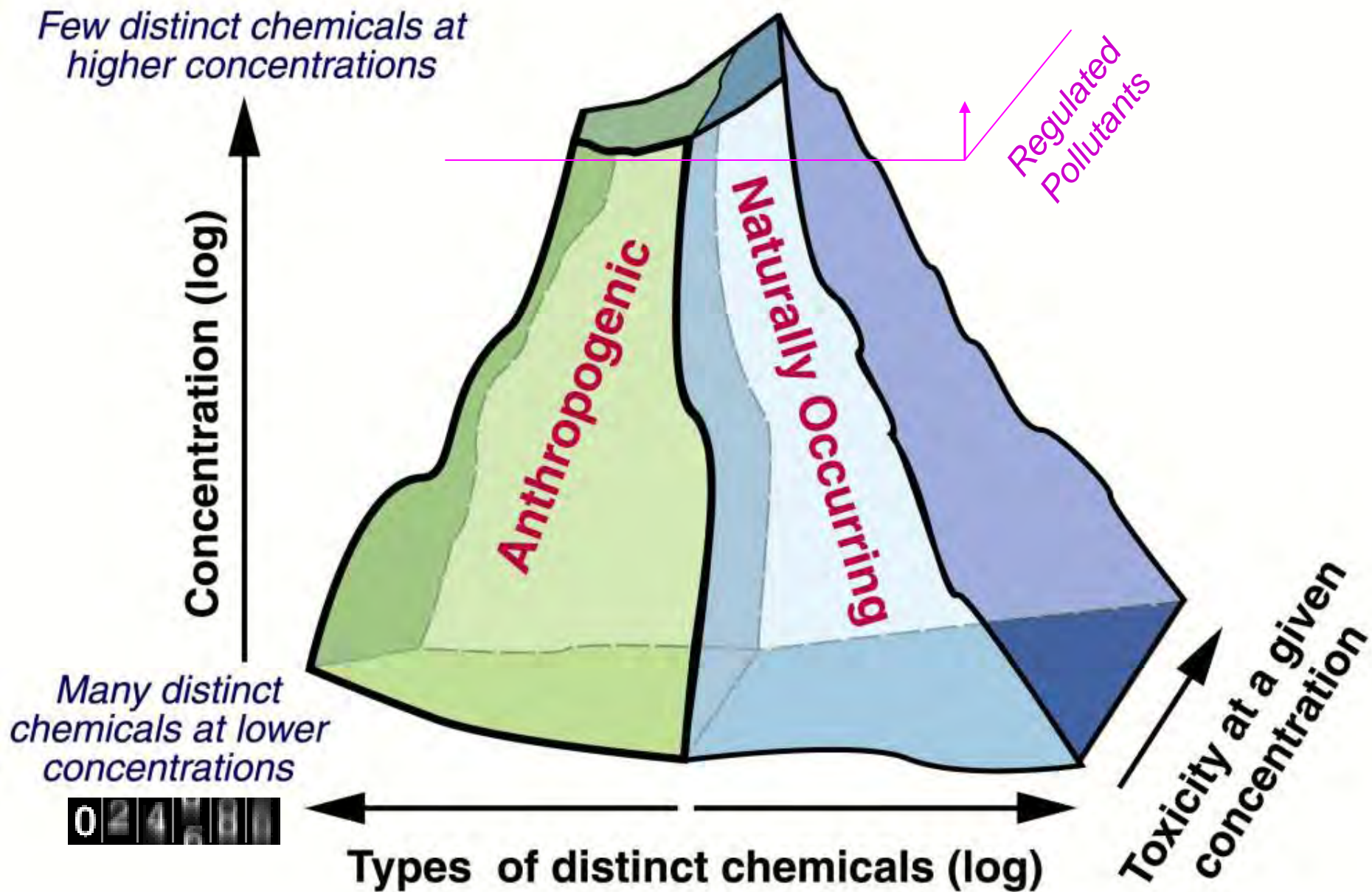
C.G. Daughton  
U.S. EPA July 2002

# Chemical Analysis Output for a Typical Environmental Sample



TICs = tentatively identified compounds

# Prevalence/Distribution of Xenobiotic Occurrence

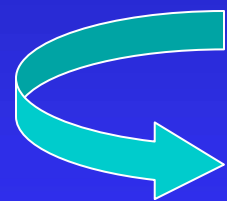


# Einstein on: *Environmental Monitoring*

“Not everything that can be counted counts,  
and not everything that counts can be  
counted.” (oft attributed to Albert Einstein)



*corollary for environmental monitoring*

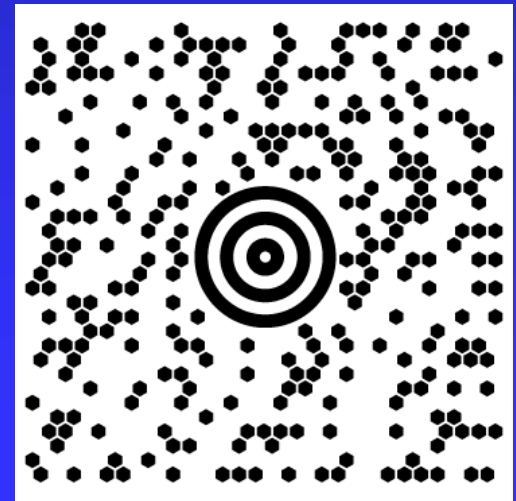


**Not everything that can be measured is  
worth measuring, and not everything  
worth measuring is measurable.**



## further truisms regarding *Environmental Monitoring*

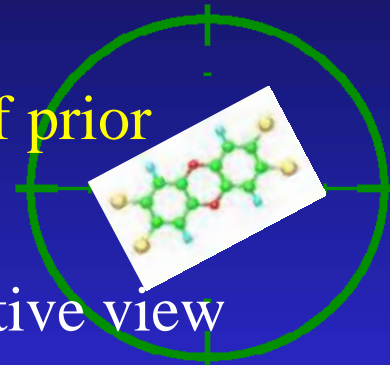
- What one finds usually depends on what one aims to search for.
- Only those compounds targeted for monitoring have the potential for being identified and quantified.
- Those compounds not targeted will elude detection.
- The spectrum of pollutants identified in any sample often represent but a small portion of those present and they are of unknown overall significance with respect to risk.



## *Biasing Our Perspective of Chemical Exposure?*

Monitoring studies tend to target those chemicals that have already been reported.

Targeted analytes are often self-selected on the basis of prior positive results.



- Are published data providing a highly biased, selective view of the world of chemical contaminants?
- How large is the chemical world that monitoring activities are overlooking?
- How do we establish how accurate and comprehensive the current picture is?
- Is our view of real-world chemical flawed?

*Are we headed in the right direction?  
What should we expect to achieve?*



- Are our priorities in balance with society's expectations and needs?
- Are we focusing on those areas that will yield the most important outcomes?
- Is research sufficiently coordinated and leveraged - internationally?

## *Possible Major Outcomes from Emerging Contaminants Research*

- Improve our ability to assess risk more comprehensively.
- Improve the public's understanding of risk.
- Expand the known universe of chemical stressors. Provide means to identify previously unrecognized or unexpected contaminants.



## *Possible Major Outcomes from Emerging Contaminants Research*

- Catalyze new paradigm for screening and regulating hazard.
- Improve speed and efficiency and reduce cost of managing, mitigating, and preventing chemical pollution.
- Develop proactive capability for early warning or prediction of newly emerging contaminants.



# Concluding Questions



- Would a more descriptive term for "unregulated" chemical contaminants be useful?
- Should such a term have clear meaning for the public and regulators?
- Is it useful to call well-established ubiquitous environmental contaminants "emerging"?
- How might progress be assessed with regard to research?

# Concluding Questions



- Can a process be designed to ensure that today's "emerging contaminants" don't persist as tomorrow's concern?
- When has a chemical "graduated" from the domain of "emerging"?
- **Can a proactive system be designed that obviates the need for the term "emerging" in the first place?**
- **Can stewardship and green chemistry programs be designed to effectively prevent the entry of ECs to the environment?**



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4-7 August 2009

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Extended abstract available:  
<http://www.epa.gov/nerlesd1/bios/daughton/Daughton-abstract-EmCon09.pdf>

Slide presentation available:  
<http://www.epa.gov/nerlesd1/bios/daughton/Daughton-slides-EmCon09.pdf>