PCBs are both legacy and emerging contaminants: Evidence for Current Manufacturing Sources of PCBs

Keri C. Hornbuckle

Dept of Civil and Environmental Engineering

IIHR-Hydroscience and Engineering

The University of Iowa

iowa superfund research program

semi-volatile PCBs: sources, exposures, toxicities

1. **PCBs: Metabolism, Genotoxicity and Gene Expression in Vivo**
   Larry Robertson, Gabriele Ludewig, Garry Buettner, Howard Glauert
2. **Oxidative Stress and PCB Exposure in Mammalian Cells**
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The

AROCLORS

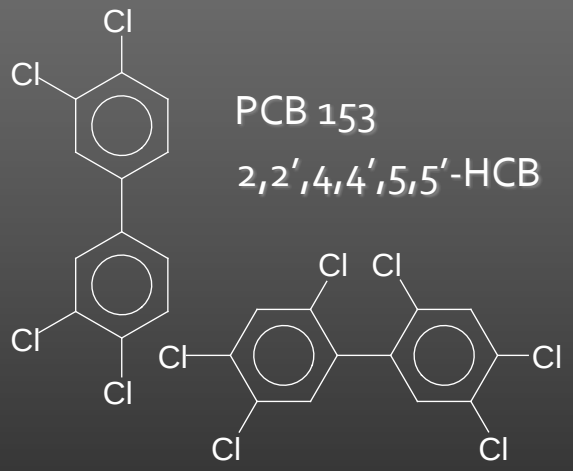
Physical Properties and Suggested Applications

APPLICATION DATA BULLETIN No. P-125

MONSANTO
CHEMICALS—PLASTICS

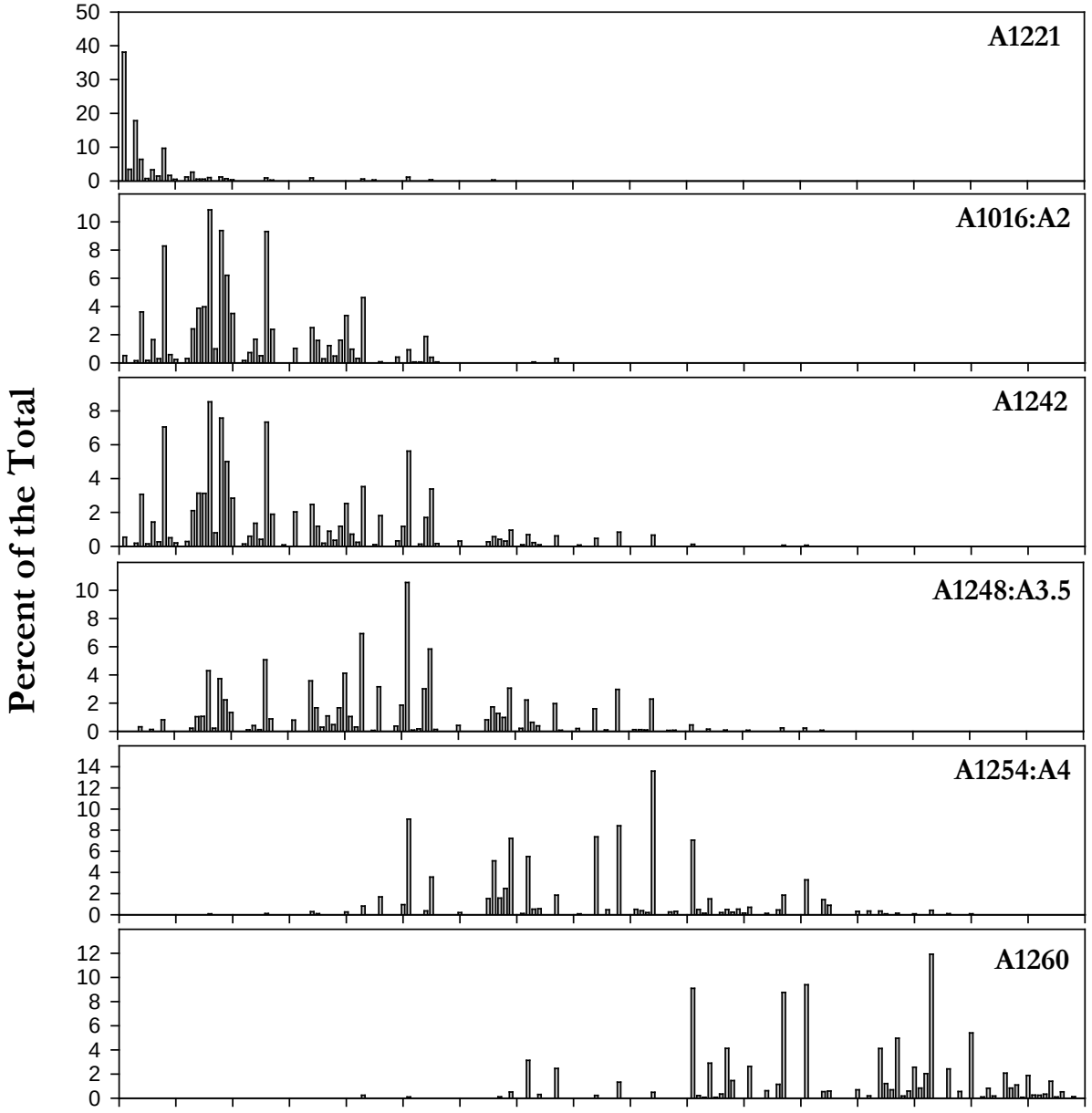
MONSANTO CHEMICAL COMPANY

Aroclors are different mixtures of PCBs



PCB 153
2,2′,4,4′,5,5′-HCB

PCB 77
3,3′,4,4′-TCB



PCBs, in order of increasing number of chlorines→

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Calculation of passive sampling rates from both native PCBs and depuration compounds in indoor and outdoor environments

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POPs
Indoor air
Sampler design

ABSTRAC

Passive sampling
and remote areas;
centrations from
passive samplers
nyls (PCBs) from
determined the sa
The uptake of nati
air and behaved a
deployed indoors
and loss of depur
dicted linear beh
behavior was ext
in *R*-values of 1.6–

Introduction

The global and regional distribution of persistent
pollutants (POPs) is now well understood, thanks in part
development of passive samplers that are lightweight, u
inexpensive and unobtrusive (Farrar et al., 2005; Gouin
2007; Kot-Wasik et al., 2007; Partyka et al., 2007).

Passive samplers that use polyurethane foam, soils, and
permeable membranes (Law et al., 1998; Migazweski
1999; Shoeib and Harner, 2002; Harner et al., 2004) have bee
world-wide and have been particularly effective in remote
across large spatial areas, and indoors where high-volum
plers (Hi-Vols) are impractical (Shoeib and Harner, 2002).

Currently, the comparison between Hi-Vol data and p
sampling data collected for specific compounds, such as P
difficult due to the difference in temporal resolution, low de
limits for Hi-Vol samples, and differences in calculating con
tions. Researchers, however, have already compared the
ences in reported calculated concentrations between Hi-Vo
passive samplers (Gouinet al., 2005a,b; Harner et al., 2006).
to Hi-Vol data and PAS-PUF data, PCB concentrations for PA
are calculated in different ways as well. Researchers derive
pling rate, or R-value ($m^3\ d^{-1}$), from either loss of depuratio

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Spatial Distribution of Airborne Polychlorinated Biphenyls in Cleveland, Ohio and Chicago, Illinois†

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Passive samplers were deployed across Cleveland, OH and Chicago, IL to evaluate the spatial variability of airborne PCBs in urban areas. We measured ΣPCB concentrations, the sum of 151 congeners or congener groups quantified using tandem mass spectrometry, spatial distributions, and congener profiles in two urban areas in the Great Lakes region. Mean ΣPCB concentrations were significantly different between Cleveland (1.73 ± 1.16 ng m^{-3}) and Chicago (1.13 ± 0.58 ng m^{-3}) during the August 2008 sampling period. Mean congener profiles were compared with commercial Aroclor mixtures and found to be similar to Aroclor 1242 in Cleveland and similar to a mixture of 1242 and 1254 in Chicago. We observed large spatial variation in concentrations and weak or no significant autocorrelation between sites in both cities. "Hot spots" of high ΣPCB concentrations were identified in both urban areas and the congener profiles at these locations were most strongly correlated to that of PCB Aroclor mixtures. Congener profiles showed important differences including the enrichment of dioxin-like congeners in Chicago.

Introduction

Urban areas are known sources of polychlorinated biphenyls (PCBs) to the regional atmosphere (*1*–*13*). Although a number of studies have examined city specific PCBs concentration, little is known about inter- and intracity variability of PCB concentrations and their emission sources. Only a few studies have examined the spatial distribution of airborne PCBs within large urban areas. Basu et al. (*14*) measured airborne PCBs at two locations in Chicago, Du et al. (*15*) measured airborne PCBs at 32 sites across Philadelphia, and Hu et al. (*16*) measured airborne PCBs at 37 sites across Chicago although they reported only the concentration of one congener (3,3'dichlorobiphenyl). Historically, PCB measurements of urban air have been made using single-site high-volume (Hi-Vols) samplers (*1*, *3*–*7*, *9*, *17*). Long-term monitoring studies of temporal trends and regional variations in airborne PCBs have also traditionally used Hi-Vols (*18*–*22*), but they do not represent spatial trends of PCBs across an urban area.

Recently, researchers have turned to passive sampling to measure the spatial distribution of PCBs. Baek et al. (*23*) and Yeo et al. (*24*) report such measurements in South Korea, Jaward et al. (*25*) in Europe, and Pozo et al. (*26*) globally with the GAPS study. Passive sampling can generate important information on the spatial distribution of PCBs across an urban area. First, "hot spots", areas of high concentrations of PCBs can be identified (*27*–*29*) and these hot spots can form the basis for further st
of PCBs and remediation e
representation of mean PC
may be estimated from man
than single-point measure
airborne PCBs in cities nea
important because urban a
major source of PCBs no
cumulating in the fish. A
consumption advisories, es
continued elevated concent

This is the first study, to
concentrations spatially dist
areas with data collected du
hypothesized that the two
Chicago, would exhibit diff
congener profiles. Our spec
to compare the spatial vari
Chicago in terms of ΣPCB co
and toxicologically relevant

Methods and Materials

Selection of Sampling Site

to select sampling sites in
required a prior knowledge
to optimize site selection
intracity PCB data do not ex
we relied on two different cri
In Cleveland, we used a no
to capture the spatial var
minimizing the number of si
we estimated the spatial dist
of airborne particles smalle
for PCB concentrations. Foll
land-use and land-cover data
emission sources for PM_{10}, p
aerosol optical depth data ta
for meteorological conditions
variance of concentrations
that our budget allowed.
variance using eq 1 as the l
relation is near 0 ($\rho_z \approx 0$) b
its neighboring sites (Z_j). In
$\mu g\ m^{-3}$. K is defined as Z_0

$$\max |Z| = \sum_{j=1}^{n}$$

In Chicago, site selection
than optimization (*16*). The
chosen based on existing a
and businesses, all in resid

† Part of the special section "Sources, Exposures, and Toxicities of PCBs in Humans and the Environment".
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Atmospheric PCB congeners across Chicago

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ABSTRACT

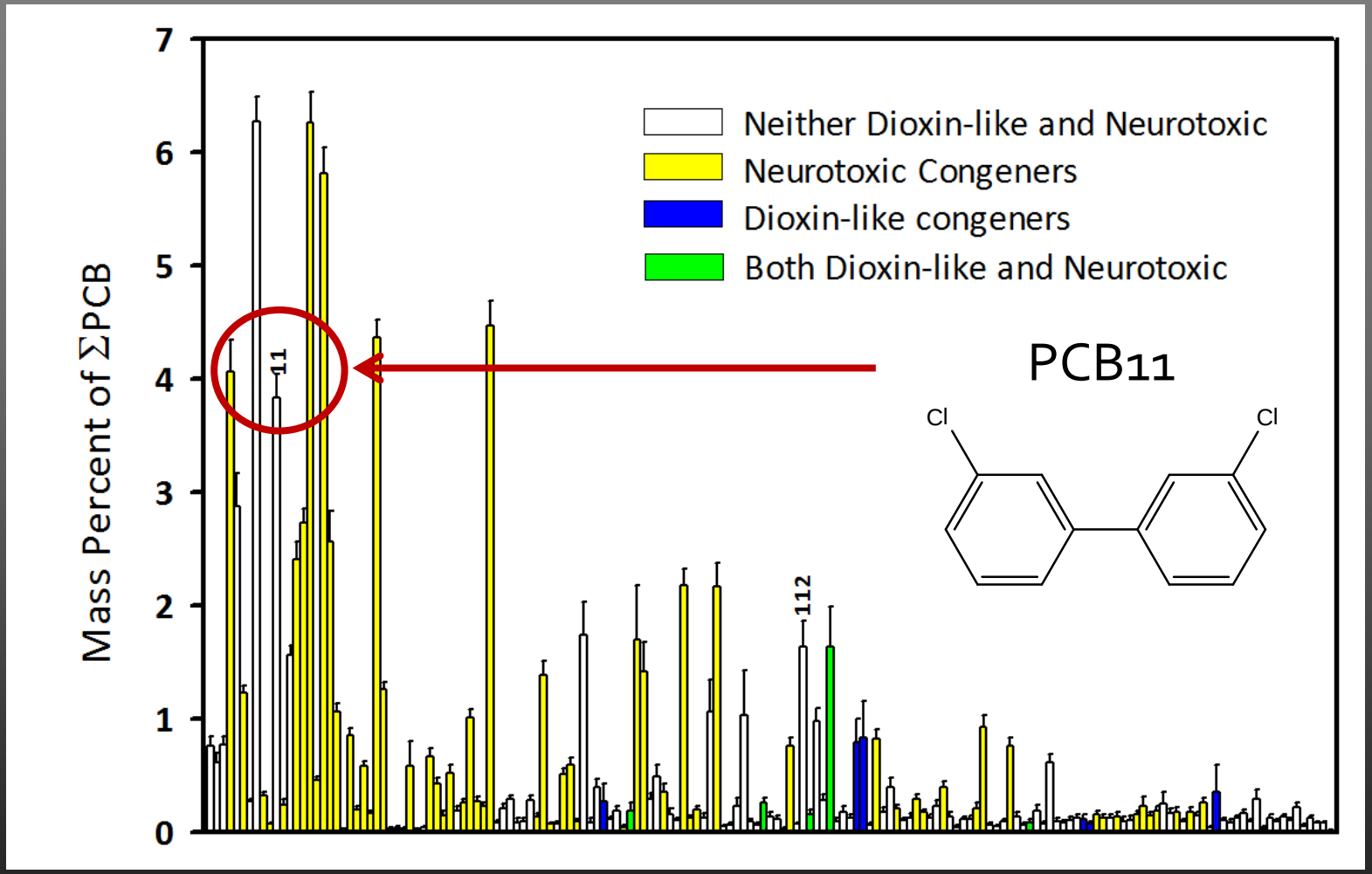
We have measured PCBs in 184 air samples collected at 37 sites in the city of Chicago using an innovative system of high-volume air samplers mounted on two health clinic vans. Here we describe results of sampling conducted from November 2006 to November 2007. The samples were analyzed for all 209 PCB congeners using a gas chromatograph with tandem mass spectrometry (GC–MS/MS). The ΣPCBs (sum of 169 peaks) in Chicago ranged from 75 pg m^{-3} to 5500 pg m^{-3} and primarily varied as a function of temperature. The congener patterns are surprisingly similar throughout the city even though the temperature-corrected concentrations vary by more than an order of magnitude. The average profile resembles a mixture of Aroclor 1242 and Aroclor 1254, and includes many congeners that have been identified as being aryl hydrocarbon receptor (AhR) agonists (dioxin-like) and/or neurotoxins. The toxic equivalence (TEQ) and neurotoxic equivalence (NEQ) in air were calculated and investigated for their spatial distribution throughout the urban-industrial complex of Chicago. The NEQ concentrations are linearly correlated with ΣPCBs while the TEQ concentrations are not predictable. The findings of this study suggest that airborne PCBs in Chicago are widely present and elevated in residential communities; there are multiple sources rather than one or a few locations of very high emissions; the emission includes congeners associated with dioxin-like and neurotoxic effects and congeners associated with unidentified sources.

1. Introduction

PCBs are a group of 209 semi-volatile anthropogenic compounds (congeners) that are commonly measured in air throughout the globe and dispersed to regions far from human activity (Ockenden

quite difficult to accurately measure the spatial distribution of PCB concentrations and hence there are very few datasets available to support current modeling and estimation methods (Breivik and Alcock, 2002; Breivik et al., 2002b). The difficulty is due to the

PCB Congener Profile in Chicago Air



PCBs, in order of increasing number of chlorines→

The University of Iowa

Environ. Sci. Technol. XXXX, xxx, 000–000

Evidence for Unique and Ubiquitous Environmental Sources of 3,3′-Dichlorobiphenyl (PCB 11)

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The non-Aroclor congener 3,3′-dichlorobiphenyl (PCB 11) has been recently detected in air, water, biota, sediment, and suspended sediment. Although it has been known since at least the 1970s that this congener is produced inadvertently during the production of diarylide yellow pigments, this work presents the first evidence that the use of these pigments in

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Passive Air Sampling of Polychlorinated Biphenyls and Organochlorine Pesticides at the Korean Arctic and Antarctic Research Stations: Implications for Long-Range Transport and Local Pollution

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…e and Engineering, Pohang …ology (POSTECH), San 31, …90-784, Republic of Korea, …ironmental Sciences, …gh, 1265 Military Trail, …1A4, Contaminant Sciences, …eries and Oceans Canada …, Sidney, British Columbia, …lar Research Institute, …gdo-Dong, Yeonsu-Gu,

and predictions of long-range transport potential. Dominance of heavy PCBs on the roof of the main building at Ny-Ålesund and a concentration gradient with distance from the main building at King George Island strongly indicated the influence of local sources. OCP levels were also influenced by long-range transport but not by local sources. This study highlights the feasibility of using passive air sampling to assess both long-range transport and local pollution in remote regions.

Introduction

Polychlorinated biphenyls (PCBs) an… pesticides (OCPs) are classified and … organic pollutants (POPs) under the S… Produced for industrial purposes, glo… emissions are relatively well establis… around Northern midlatitude reg… ubiquitous, being detected regularly … polar regions. The occurrence in thes… has been explained by the process … and fractionation (*4*), and long-range … (LRAT) has been identified as one of … POPs to polar regions (*5*−*7*). The … influence of anthropogenic activities … is illustrated by reports of elevat… atmosphere and sediments at McMu… Antarctica (*8*, *9*). Antarctic soils we… contaminated by remote and local so… se studies suggest that both LRAT a…

Detecting a new PCB in Chicago air

Although banned some 30 years ago, PCB compounds are being discovered in urban ambient air. Non-Aroclor PCB (3,3′-dichlorobiphenyl), or PCB11, discovered in a recent study published in *ES&T* (**2008**, *42* [21], 7873−7877), is considered an "inadvertent PCB," says Keri Hornbuckle of the University of Iowa, a coauthor of the new study. PCB11 was not one of the congeners manufactured by Monsanto Chemical Co., and it has been regularly reported as nondetectable in Monsanto's mixtures, Hornbuckle says.

"I think it is a tremendous finding," says Bob Herrick of the department of environmental health at Harvard University. "Most people have never seen anything published about this congener," he adds. "The paper shows evidence of something that we didn't see [before]," says Ann Casey, senior chemist with Northeast Analytical, Inc.

PCBs are a set of 209 congeners. Of that total, about 150 were sold by Monsanto as components of Aroclors. As much as 1400 million pounds of PCBs was produced as Aroclor mixtures from 1930 to 1970, says Casey. She estimates that 1233 million pounds of the Aroclor PCBs was used in the U.S. before they were banned in 1977.

Hornbuckle and her colleagues collected air samples in Chicago from November 2006 to November 2007 by using vehicle-

levels: the concentration of PCB11 was as much as 15% of measured PCBs in the air. The concentrations also varied by season (and temperature), following a typical trend of PCB levels found in ambient air in urban and remote settings.

TIM SEVEN

Hornbuckle checks an air sampler perched on top of a medical clinic van on a street in Chicago.

What makes this congener inadvertent is that it appears to have been created by weathering, volatilization, or dechlorination and may not be related to Aroclor mixtures, Hornbuckle says. She and her colleagues aren't sure what the source is, but they theorize that the PCB11

such plants in and near the city. Hornb… The … with ga… spectro… analyti… scienti… mologi… measu… Inde… tute fo… Health… nanogr… Aroclo… Casey … measu… per cu… GC/… tion as… has no… search, … Keri's … the for… The … that re… yond t… studyin… researc… "Wh… have t… and n… ture," … burg, … environ… ter for … at Rutg… PCB… there, … sitivity …

Research

Partial Pressures of PCB-11 in Air from Several Great Lakes Sites

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School of Public and Environmental Affairs, Indiana University, Bloomington, Indiana 47405

Revised manuscript received July … 2009.

…ons of 3,3′-dichlorobiphenyl (PCB-… …ater and air, and it has been suggested … come from commercial PCB … ata on atmospheric partial pressures … ls at five sites around the Great … t both total PCBs and PCB-11 track … In addition, we show that, with … he PCB congener pattern at our … sented by Aroclor 1242; in Chicago, … nted by a 1:1 mixture of Aroclors … ssible exception of Sturgeon Point, … B-11 in air have not changed … December 2007. The source of PCB-… v pigments, to the degradation of … B congeners, or to both.

…ls (PCBs) have been of intense … r about 40 years. These compounds … ures of various homologues and … ed congeners) and marketed for a … ginning in about 1935 and ending … es. Aroclor was the most common … e PCB mixtures. It turned out that … ly persistent, and their production, … ed States were banned, although … were allowed to remain "in use," … nsformers and capacitors. … rcial PCB mixtures, such as the … r) the most significant sources of … individual PCB congeners can also … cts during the production of other … leased to the environment. These … ls" include 3,3′-dichlorobiphenyl … ne from the production of dichlo… view, see ref *1*. Perhaps as a result … as been found in New York/New … Delaware River water (*3*), in Arctic … hicago air (*5*), and in Philadelphia … ponent of the commercial Aroclor … United States or in the Clophen … in Europe (*7*, *8*).

Chicago, as part of the Integrated Atm… Network (IADN) (*9*, *10*). We added PCB… starting in January 2004. Two of our st… might be expected to have relatively … use of industrial chemicals, including … sites are located in sparsely populated … any use of industrial chemicals is ex… were a major source of PCB-11, we … concentrations of this compound wou… sites and lower at remote sites. We w… detailed data on PCB-11 atmosphe… obtained in Chicago with those publi…

Experimental Section

Sampling Sites. In this paper, data … samples collected at the five United St… IADN sites from January 2004 until … remote sites are Eagle Harbor on Lake … Bear Dunes on Lake Michigan. The u… on Lake Michigan and Cleveland on L… Sturgeon Point, on Lake Erie, is within … York, and represents a rural site. A m… sites is given in Figure 1.

Sampling Methods. Air samples a… days using a Graseby (General Metal … high volume air sampler fitted with a … filter (QM-A) followed by an Amberlite … (Supelco) cartridge to trap particle ar… respectively. The samplers are calibrat… air for 24 h to get a total sample volu…

Sample Extraction and Cleanup. … Soxhlet extracted using 50% by volu… (OmniSolv, EM Science) for 24 h. Sele… 14, 65, 166) are added as surrogate re… raw extracts are then concentrated b… solvent exchanged to hexane, and fr… water deactivated silica gel (Aldrich Ch… 634, 100−200 mesh) column using hexa… dichloromethane in hexane. PCBs … fraction. The cleaned up samples are … by rotary evaporation and N_2 blown-… congeners 30 and 204 are added as i… instrumental analysis (*12*).

Instrumental Analysis. A total of … measured by an Agilent 6890 gas chro… with an Agilent 7683 autosampler. Th… separated on a DB-5 60 m column (25… film thickness) and analyzed using … capture detector (ECD). The carrier gas … makeup gas is N_2. PCB peaks are iden… times compared to a calibration sta… separated from other PCBs, and there… ences. The identity of this congener … 60 m 1701 column (250 μm i.d. and 0… The Supporting Information shows ex… grams of atmospheric extracts and aut… using both DB-5 and 1701 GC liquid p… done using an internal standard meth…

Research

Discovery of Non-Aroclor PCB (3,3′-Dichlorobiphenyl) in Chicago Air

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Air samples were collected in Chicago, Illinois in 2007, and 3,3′-dichlorobiphenyl (PCB11, CAS 2050-67-1) was detected and quantified using GC/MS/MS in 91% of 184 samples. To the best of our knowledge, this is the first published report of PCB11 in ambient air. This compound is ubiquitous in air throughout the city of Chicago. The annual mean concentration in air samples collected from November 2006 to November 2007 is 24 pg m^{-3} (±24 pg m^{-3} SD), although the seasonal variation is significant. The concentration of PCB11 is up to 15% of measured polychlorinated biphenyls (PCBs) in air but only up to 0.16% of commercial Aroclor mixtures that were banned from production in the 1970s. PCB11 is associated with pigments, paints, and resins and has been reported to be a dominant congener among PCBs detected in the wastewater effluent from paint production. The wide distribution of PCB11 in Chicago air is consistent with volatilization of this compound from painted surfaces although the actual source of PCB11 is unknown.

Introduction

important impacts on regulatory decisions in the future, although its source, toxicity, and potential for human exposure need further investigation.

Materials and Methods

Air Sampling. Air was sampled using high-volume air samplers (HI-Vols) equipped with quartz fiber filters and XAD-2 resins. HI-Vols were mounted on platforms attached to the rear of two medical clinic vans (Figure 2). The HI-Vol platforms were designed to raise the sampler to the top of the van for operation and lower the sampler for filter and XAD replacement.

The sampling locations (Table S1) were primarily elementary schools where the mobile clinics provide service to the students and their families for diagnosis and treatment of asthma and related respiratory illness. When the vans visited the schools for clinical service, the HI-Vols collected air samples for the 6−8 h period that the van remained at the school. The samplers were operated with the assistance of the trained staff at Mobile C.A.R.E. Foundation of Chicago (Comprehensive Care for Chicagoland's Children with Asthma). Both vans went out for clinical service usually on the same day so two samples were collected at two different sites on most sampling days. The air was pulled with a vacuum pump through a quartz fiber filter to retain particles and then through an XAD-2 resin cartridge to collect PCBs in the gas phase. Temperature and relative humidity were recorded every 5 min on one van by the CR800 measurement and control system (Campbell scientific, Inc., Logan, UT). The flow pressure drop was recorded daily using a magnehelic pressure gauge mounted on each sampler.

Sample Analysis. After collection, all samples were placed in sealed ziplock bags and stored in a refrigerator until shipped to the laboratory. Once the samples were transported to the laboratory, they were logged and stored at 4 °C until extracted. The filter and XAD were extracted by accelerated solvent extraction (ASE 300, Dionex, Sunnyvale, CA) using an optimized EPA Method 3545 (*11*). The extraction cell was preheated for 5 min to 100 °C, followed by a static extraction

Summary of the Clues about PCB11:

1. Not present or trace in commercial Aroclor mixtures
2. Detected in the wastewater effluent from paint production (Litten et al., 2002, *Chemosphere*)
3. Widely distributed in the urban air of Chicago and Cleveland
4. Global pollutant
5. Concentration proportional to population
6. Present in many consumer products
7. Structural similarity of with some pigment intermediates
8. No direct significant emission from facilities

Hypothesis:

The source of airborne PCB11 is painted surfaces (paint).

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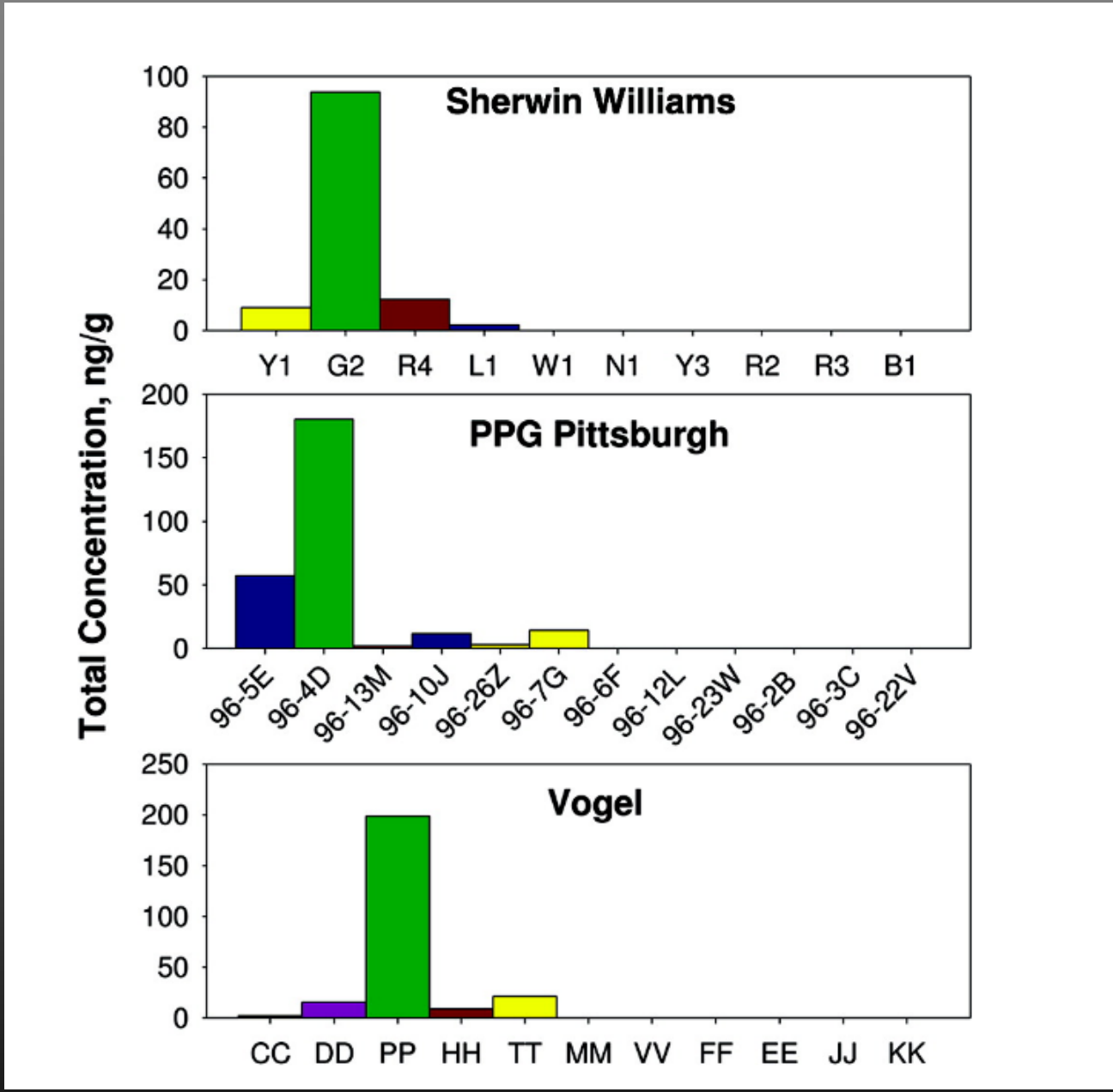
Paint Bases contained no PCBs

Inorganic pigments contained no PCBs

- Titanium dioxides: white
- Iron oxide: yellow, orange, red, brown and black
- Carbon black
- Umber
- Sienna
- Ochre

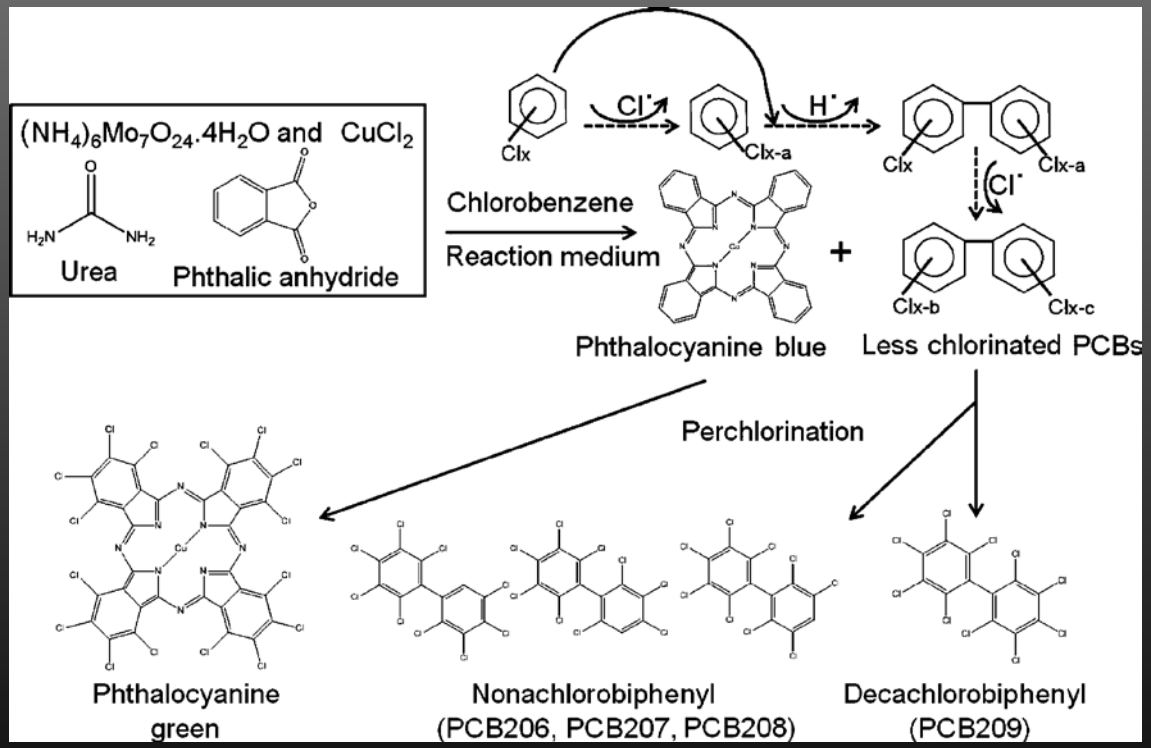
Organic Pigments contain PCBs



Hu and Hornbuckle, 2010 ES&T

Organic pigments

- **Azo** pigments: yellow, orange, red, brown
- **Phthalocyanine** pigments: blue and green
- Polycyclic pigments
- Heterocyclic pigments



Environ. Sci. Technol. 2010, 44, 2822–2827

Inadvertent Polychlorinated Biphenyls in Commercial Paint Pigments†

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A polychlorinated biphenyl (PCB) that was not produced as part of the Aroclor mixtures banned in the 1980s was recently reported in air samples collected in Chicago, Philadelphia, the Arctic, and several sites around the Great Lakes. In Chicago, the congener 3,3′-dichlorobiphenyl or PCB11 was found to be the fifth most concentrated congener and ubiquitous throughout the city. The congener exhibited strong seasonal concentration trends that suggest volatilization of this compound from common outdoor surfaces. Due to these findings and also the compound's presence in waters that received waste from paint manufacturing facilities, we hypothesized that PCB11 may be present in current commercial paint. In this study we measured PCBs in paint sold on the current retail market. We tested 33 commercial paint pigments purchased from three local paint stores. The pigment samples were analyzed for all 209 PCB congeners using gas chromatography with tandem mass spectrometry (GC-MS/MS). More than 50 PCB congeners including several dioxin-like PCBs were detected, and the PCB profiles varied due to different types of pigments and different manufacturing processes. PCB congeners were detected in azo and phthalocyanine pigments which are commonly used in paint but also in inks, textiles, paper, cosmetics, leather, plastics, food and other materials. Our findings suggest several possible mechanisms for the inadvertent production of specific PCB congeners during the manufacturing of paint pigments.

Some PCB congeners
are not present or are
unfavored or improbab
manufacturing process (2
PCB congeners. In air s
2007, we found PCB11
city (4). The compound w
in air of polar regions (5)
reported in air of Philade
Great Lakes (7). It appea
In addition, PCB11 was m
from paint production. P
dechlorination is not lik
are in very low conce
widespread distribution

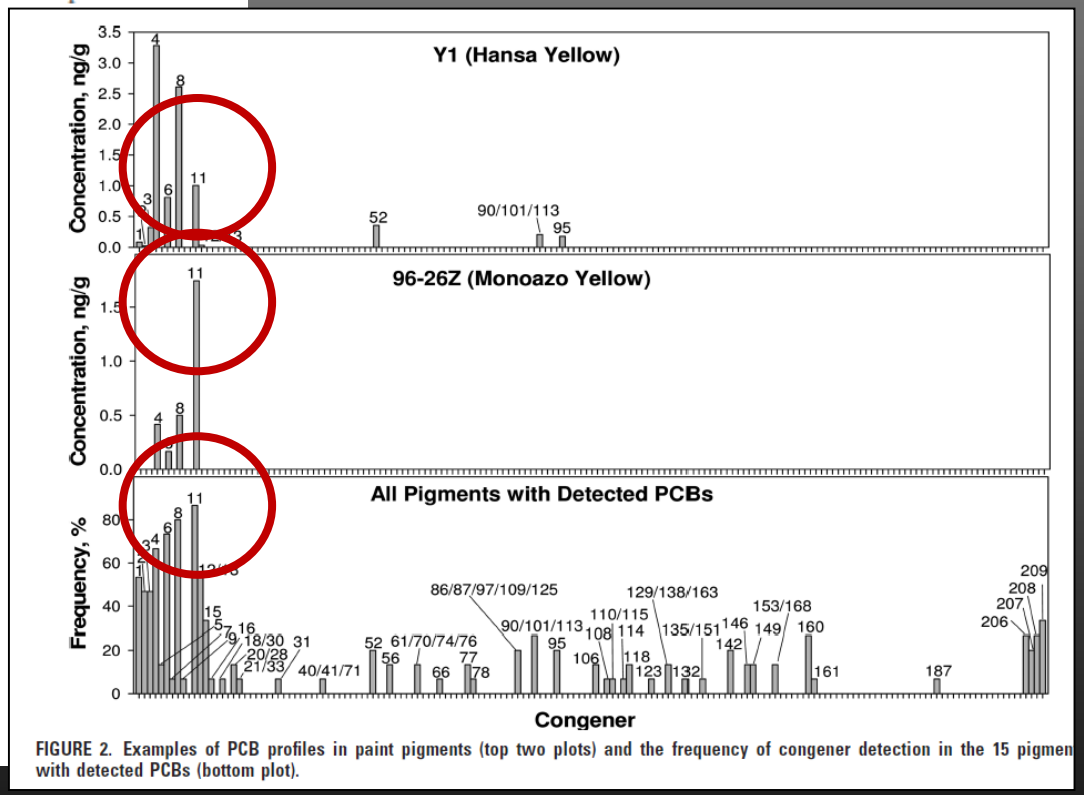

FIGURE 2. Examples of PCB profiles in paint pigments (top two plots) and the frequency of congener detection in the 15 pigmen with detected PCBs (bottom plot).

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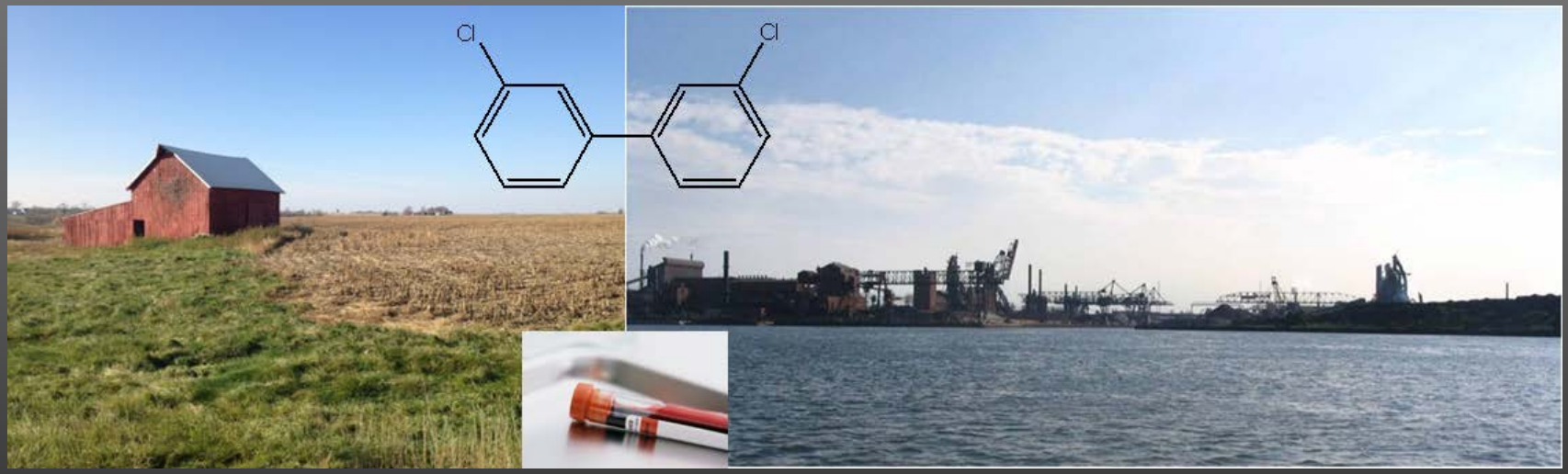
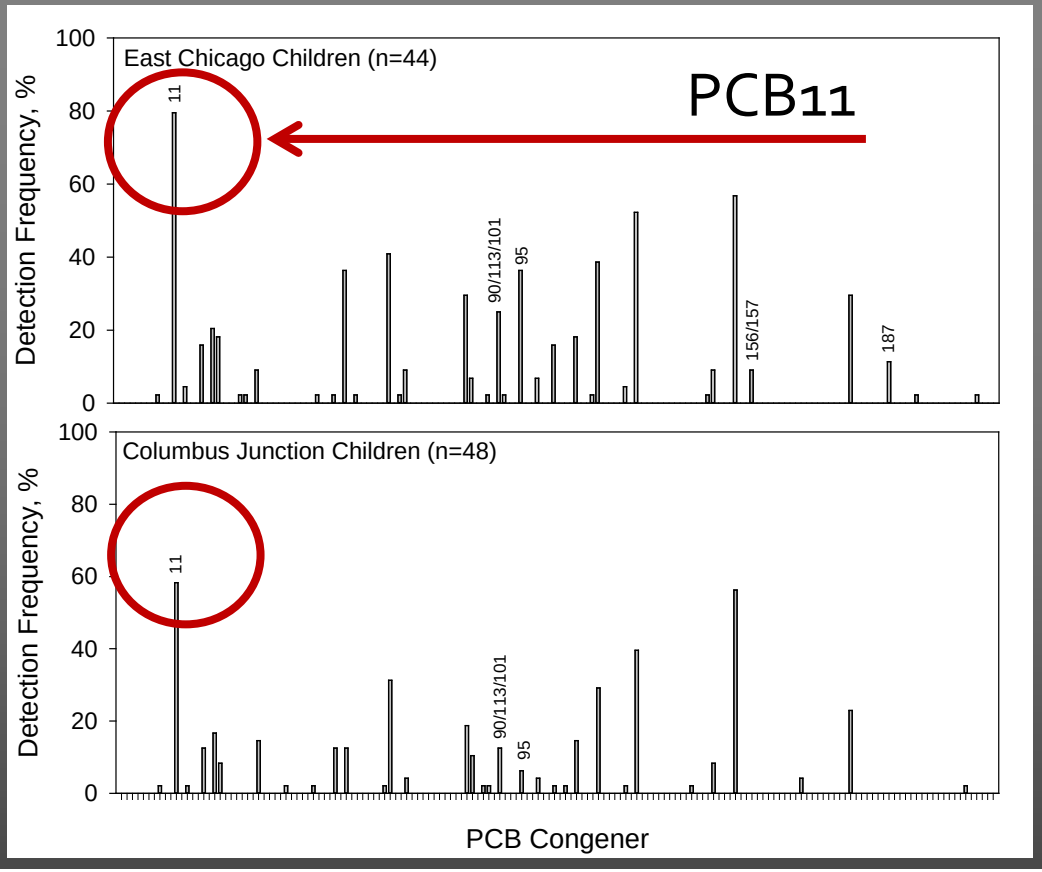
PCBs are in human blood serum, including PCB11



PCB congeners detected in children and their mothers



Environmental Science & Technology

2013

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PCBs and OH-PCBs in Serum from Children and Mothers in Urban and Rural U.S. Communities

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Supporting Information

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