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Polychlorinated Biphenyls (PCBs) in General Consumer Products

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This report is available on the Department of Ecology's website at
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Department of Ecology
Olympia, Washington

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Introduction

In this study, the Washington State Department of Ecology (Ecology) evaluated the presence of polychlorinated biphenyls (PCBs) in general consumer products. Particular emphasis was placed on products likely to be contaminated with PCBs due to the inadvertent production of PCBs in the manufacturing process. Although many chemicals may contain low levels of PCB contaminants due to use of chlorine in their manufacture, recent studies have shown PCB presence in pigments and dyes used in consumer products. Products known to contain PCB contaminants include paints (Hu, 2010), newspapers, glossy magazines, cereal boxes, yellow plastic bags, etc. (Rodenburg, 2012).

All PCBs are of interest but four specific PCBs (PCB-11, -206, -208, and -209) were selected for the study. Historically, these four PCB congeners are not a major constituent in Aroclor mixtures and are believed to be the result of inadvertent PCB creation during the manufacture of pigments and dyes. The goals of the study were to 1) evaluate the level of PCBs in various consumer products, and 2) confirm the continued presence and potential environmental release of PCBs within Washington State. This report summarizes the results of the four specific congeners of interest.

In this study, 68 products were tested for PCBs. Several products were separated into more than one sample and 74 samples were submitted for analysis. PCB-11 was found in 66% of the samples tested. An additional 2% of the samples are estimated to be near the reporting limit and are likely to contain PCB-11.

PCB-206 and -208 were not present in most of the products tested. One sample contained both PCB-206 and -208 while seven contained PCB-209. One product, a phthalocyanine green-based colorant used to color white paint, contained detectable levels of all four PCBs and is the only sample to contain PCB-206 and -208 at detectable levels.

The samples were separated into product categories including packaging, paper products, paints and paint colorant, caulks and a miscellaneous category consisting of two printer inks and two food samples. PCB-11 was found in all product categories in the range of 1 to 45 parts per billion (ppb). These results confirm the presence of PCB contamination in consumer products.

Almost all paint and colorant samples tested contained one or more PCBs at detectable levels. Packaging and paper products sampled contained PCBs, particularly PCB-11. Currently, caulks for sale do not appear to be a significant PCB source. PCB-11 was found in one caulk that changed from pink to white during curing. Given the small sample size, further evaluation of caulks is warranted.

Additional testing of other products such as clothing, cosmetics, soaps and hand sanitizers and household cleaning products is needed to evaluate other potential PCB sources. Products containing yellow, green and white pigments particularly warrant additional sampling.

Based upon the results of this study, Ecology concludes that PCBs:

- Are found in consumer products.
- Can enter the environment in significant concentrations through water and air discharges.

- May affect people directly through contact with consumer products.

Please note that this report does not investigate the effects of PCBs on human health or the environment, nor does it reach any conclusions concerning the risk they pose.

Background

Polychlorinated biphenyls (PCBs) are a class of persistent, bioaccumulative, and toxic (PBT) compounds that historically had a wide range of uses, including consumer products. PCBs are created by reacting biphenyl with chlorine (Pomerantz, 1978). PCBs were used in:

- Electrical transformers and capacitors
- Heat transfer and hydraulic systems
- Vacuum pumps and lubricants
- Surface coatings
- Adhesives
- Plasticizers
- Inks
- Insulating materials
- Pesticides

(UNEP, 2007)

From 1929 to 1979, PCB production in the United States was approximately 1.4 billion pounds (600,000 metric tons), with 77 percent used in transformers or capacitors (EPA, 1976). PCBs were valued for their persistence, inability to conduct electricity, flame retardancy, plasticizing, and anti-microbial effects. Commercial PCB production ended by 1979 under the Toxics Substances Control Act (TSCA), but inadvertent PCB generation continues. Current levels of PCBs in Washington stem from cycling of PCBs in the environment, continuing releases from historic uses, and releases of newly generated PCBs.

In the Puget Sound, surface runoff is the largest pathway to aquatic environments, followed by wastewater treatment plants, and air deposition. PCBs are released in the highest quantities in commercial areas compared to other land covers, making PCB contamination especially relevant to the highly urbanized Puget Sound Basin (Ecology, 2011).

Studies indicate that PCBs are ubiquitous throughout the natural environment, in air, soil, and sediments, and are found in animals throughout the food chain (ATSDR, 2000). PCBs were detected in migrant Chinook salmon tissue and fish, and other marine mammals locally important to the Puget Sound region. Concentrations, however, appear to be declining in Puget Sound harbor seals and mussels. No equivalent trend can be identified in fish, although modeling suggests levels will start to decline in English sole by 2020 (Ecology, 2011). Fish consumption advisories have been issued for both marine and fresh water species in Washington due to PCB concentrations.

Historically, PCBs were manufactured in nine major mixtures called Aroclors. Aroclor was the tradename of the technical mixture of PCBs sold in the United States by Monsanto Chemical Company (Monsanto). Prior to 1979, Monsanto in Sauget, Illinois produced approximately 99% of the PCBs used within the U.S. The nine Aroclor mixtures included Aroclors 1016, 1221, 1232, 1242, 1248, 1254, 1260,

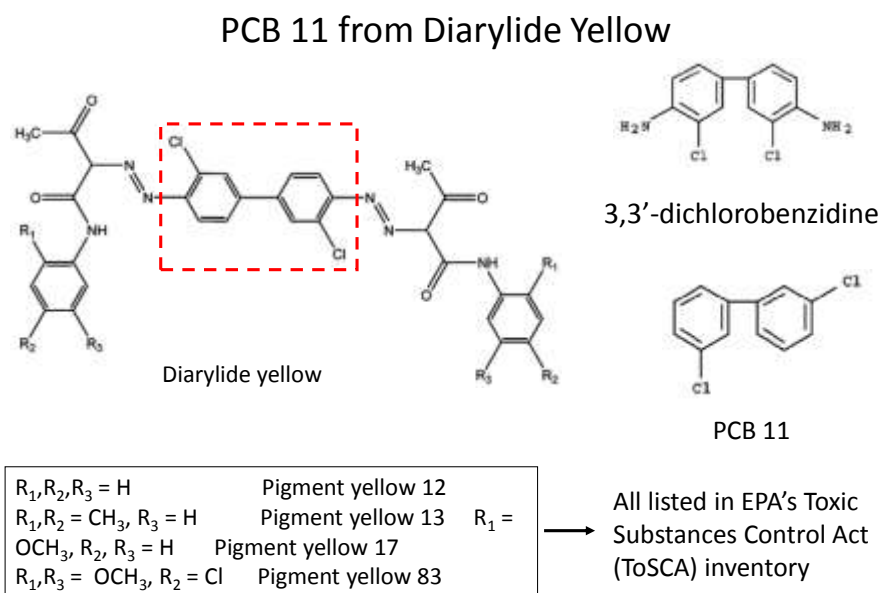
1262 and 1268 (ATSDR, 2000). Production of these Aroclor mixtures decreased from over 86 million pounds in 1970 to 35 million pounds in 1977 (EPA, 2012a).

PCBs in Products

PCBs can still be found in products (Hu, 2010; Rodenburg, 2012). Many of these products contain PCBs as an impurity created during production processes. As part of rulemaking on inadvertently generated PCBs, EPA identified 200 chemical processes with a potential for generating PCBs and narrowed it to 70 with a high potential. (NYAS, 2005). Hu et al. (2010) sampled consumer paints containing specific azo (yellow and orange) and phthalocyanine (blue and green) organic pigments and found PCB levels ranging from 2 to 200 ppb in 15 of 33 consumer paints tested. Rodenburg et al. detected PCBs in consumer products in the range of 1 to 38 ppb (Rodenburg, 2012).

Diarylide yellow comprises approximately 25% of the 250 million tons of organic pigments produced yearly worldwide (Rodenberg, 2012) and testing has shown PCBs (and especially PCB-11) are produced during its manufacture. As shown in Figure 1, PCB-11, indicated in the red box, is part of the structure of diarylide yellow. PCB-11 is either produced as a byproduct during the manufacturing process or results from degradation of the pigment.

Figure 1: Diarylide yellow and PCB-11 (From Rodenburg, 2012)



(Basu et al. 2009)

In addition to PCB-11, higher molecular weight PCBs (PCB-206, PCB-208, and PCB-209) are produced as byproducts from one of the common manufacturing processes of the inorganic pigment titanium dioxide (TiO_2) (Rodenburg, 2012). Chlorine is reacted at high temperatures with titanium dioxide (TiO_2)

ores to form the liquid titanium tetrachloride (TiCl_4). TiCl_4 is reacted with oxygen to make pure TiO_2 (UNEP, 2007). Higher molecular weight PCBs are created as a reaction byproduct.

Awareness of these issues is increasing.

- EPA recently announced the settlement of enforcement against Titanium Metals Corporation (TIMET) for the improper disposal of PCB-contaminated waste generated during titanium dioxide production. As part of the settlement, TIMET agreed to pay a \$13,750,000 penalty (EPA, 2014).
- The Japanese Ministry of Economy, Trade and Industry along with two related ministries reanalyzed 242 organic pigments found to contain PCBs as contaminants. All pigments were found to contain PCBs and 101 pigment contained PCBs over 0.5 parts per million (ppm) (METI, 2013a).
- Reanalysis of four yellow pigment samples detected PCB concentrations in the range of 59 to 1,000 ppm (METI, 2013b). Alternatives to PCB-contaminated pigments and dyes are not readily available especially in inks and paints for printing. New pigment development is a long-term project (Christie, 2014).

In addition to PCB- contamination in pigments and dyes, PCBs were used extensively in caulking compounds. It is unknown if new caulk contains PCBs. Historically, PCBs were added to joint sealant caulks at high levels to improve their flexibility, increase their resistance to mechanical erosion, and improve adherence to other building materials (Diamond *et al.*, 2010). PCBs can be lost from caulk through volatilization, as well as wash-off and erosion. PCBs in caulk are associated with higher levels of PCBs in indoor air and dust, and the external soil (Priha *et al.*, 2005; Herrick, 2007; SAIC, 2011). Larger amounts of PCBs may be released during renovations or destruction. Certain removal practices can reduce the amount of PCBs released to workers and the environment (Sundahl *et al.* 1999).

Sealants with high levels of PCBs have been found at varying levels in buildings from about 1950 to 1980 in several studies in the U.S. and other countries. In general, PCBs in caulk ranged from 5-30% (Priha *et al.*, 2005). The most comprehensive study of legacy caulk in buildings was conducted in Switzerland (Kohler *et al.*, 2005). In this study, 1,348 caulk samples from concrete buildings built between 1950 and 1980 were analyzed for PCBs. Forty-eight percent of the caulk samples contained PCBs, from < 50 ppm up to 550,000 ppm (55%). Similar results were found in smaller studies, including the 2011 study in the Duwamish (SAIC, 2011). Eight of 17 (47%) composite caulk samples from representative buildings (industrial buildings from 1950-1977) had PCB concentrations from 3 to 920 mg/kg.

Project Description

Ecology conducted a study of current consumer products that are believed to contain PCBs as a production impurity at the ppb level. The study objectives were to:

- Assess the levels of PCBs in general consumer products.
- Confirm the continued presence and potential release of PCBs to the environment in Washington.

Products were purchased based upon previous studies showing a presence of PCBs. Current products found to contain PCBs in existing studies (Hu, 2010; Rodenburg, 2012) include:

- PCBs as a contaminant (ppb level):
 - Paint
 - Glossy magazines
 - Newspapers
 - Cardboard
 - Yellow plastic bags

As indicated previously, historical caulk contained appreciable levels of PCBs, typically as an active ingredient (ppm level). Caulks currently on the market were sampled to determine if PCBs in caulk remains an issue.

Additional sources were searched to identify potential products containing PCB contamination. Sources included Material Safety Data Sheets (MSDS) for specific products, product labels, National Institute of Health's Household Product Database, sampling reports from authoritative bodies, etc.

Emphasis was placed on:

- Yellow, green, or blue products using organic azo pigments or dyes.
- White products containing titanium dioxide.

Those products containing the highest levels of pigments or dyes were prioritized. For example, product MSDSs that listed high levels of TiO_2 had priority over samples reported to contain lower levels. Product samples were sent to a contract laboratory for analysis. Due to time constraints, historical caulks were not tested although caulks currently for sale were sampled.

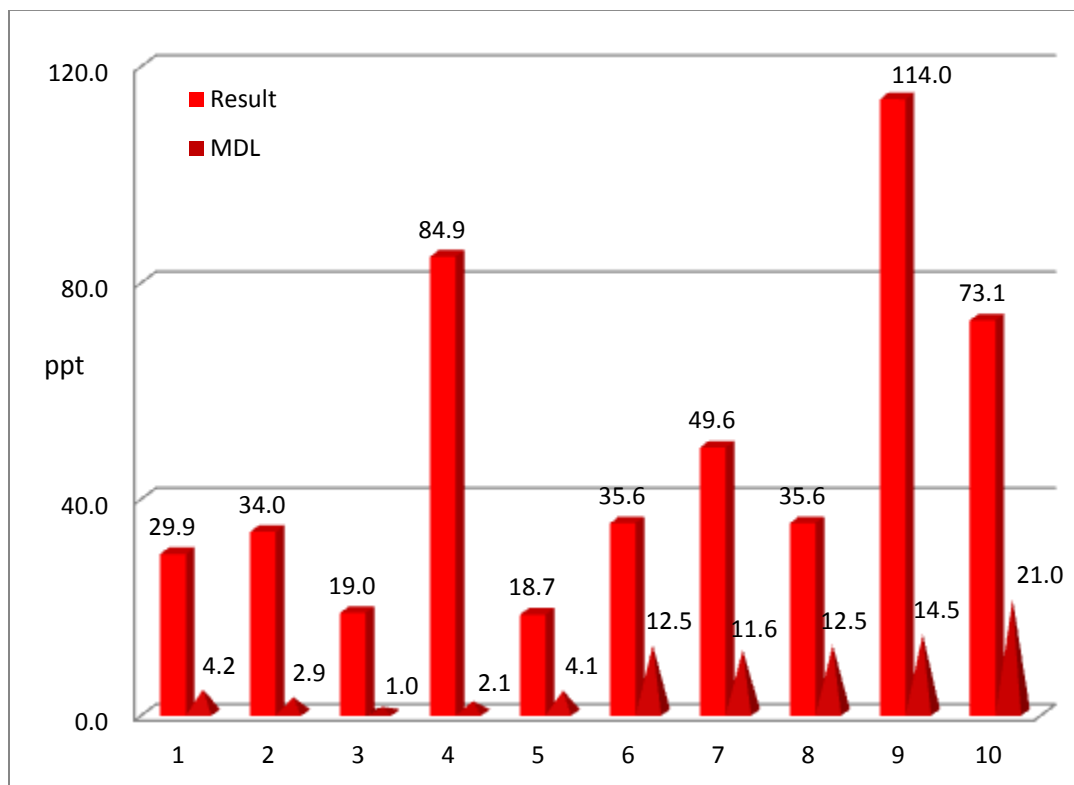
Data Quality

All samples were analyzed using EPA Method 1668A, Chlorinated Biphenyl Congeners in Water, Soil, Sediment, Biosolids, and Tissue by HRGC/HRMS (EPA, 2010). No samples were analyzed using EPA SW-846 Method 8082A (EPA, 1996) as indicated in the Quality Assurance Project Plan (QAPP).

Because of the low detection limit of these analyses at the parts per trillion (ppt) levels, the method blanks were reviewed closely. The detection limits and quantitation levels using EPA 1668 are usually

dependent on the levels of interferences and laboratory background levels rather than instrumental limitations. Ten method blanks were run during analysis of the 74 samples. Each of the method blanks reported detectable levels of PCB-11 above the method detection limit (MDL) (Figure 2) at the ppt level but below the method reporting limit (MRL). PCB presence in all the method blanks is due to background laboratory levels, which are likely from low-level contamination from equipment used to analyze PCB in previous studies.

Figure 2: Detectable amounts of PCB-11 in ten method blanks



The blank results were evaluated numerically (Table 1) and PCB-11 was found in the method blanks at an average of 49.4 ppt.

Table 1: Evaluation of PCB-11 data in method blanks

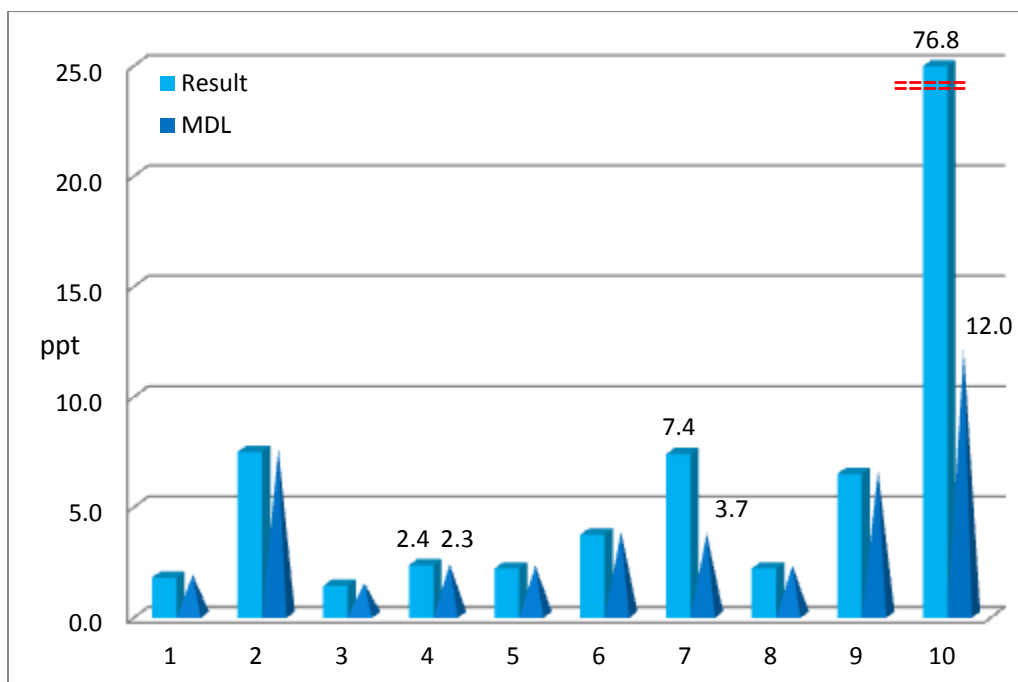
	Results	MRL	MDL
	(ppt)		
Minimum	18.7	189.0	1.0
Maximum	114.0	1,000.0	21.0
Average	49.4	307.1	8.6
Median	35.6	198.0	7.9

The presence of PCB-11 in the method blanks was a factor in establishing PCB-11 MRLs and explains the higher MRL values.

A similar evaluation was done for all the remaining PCBs in the method blanks. PCB-206 was not found in any of the blanks. PCB-208 was found in only one of the nine blanks at a value just above the MDL (4.7 ppb in Blank #4 compared with a MDL of 3.5 for the same analysis). Reported values for PCB-206 and -208 are unaffected by potential laboratory background.

PCB-209 was found in three blanks, with one blank containing an appreciable amount (Figure 3). In instances where PCB-209 was “not detected,” the reporting value was assigned the MDL. Therefore the remaining seven blanks show the same value for the sample (column) as the MDL (spike).

Figure 3: PCB-209 in ten method blanks



Evaluation of the PCB-209 blank results (Table 2) indicates that the average sampling results was 11.22 ppt and the MRL average of 768 reflects this greater variability.

Table 2: Evaluation of PCB-209 data in method blanks

	Results	MRL	MDL
	(ppt)		
Minimum	1.5	472.0	1.5
Maximum	76.8	2,500.0	12.0
Average	11.2	768.1	4.4
Median	3.1	495.0	3.0

The large outlier for sample 10 remains a concern and will be factored into the discussion of the PCB-209 results. For the purposes of this study, it will be assumed that any estimated value for PCB-11 and -209 equal to or less than 0.1 ppb (100 ppt) may potentially be due to laboratory background and is not

suggestive of PCB presence in these products. Due to no background values in the blank samples, any PCB-206 and -208 above the MDL will be suggestive of PCB presence in these samples.

All remaining QA/QC requirements established within the QAPP (Ecology, 2013) were met.

Results

Sixty-eight products were collected for testing and 74 samples were prepared for analysis (Table 3). Some samples were separated into multiple components:

- A Sunday edition of a local newspaper was separated into four individual samples:
 - Front page
 - Glossy inserts
 - Comics section
 - Non-glossy inserts
- Two mustard products were separated into the containers and contents.
- A package of white printer paper was separated into samples of the white paper and the packaging surrounding the paper.

HWTR staff purchased products from local retailers. Pictures were taken of all products prepared for analysis. Products were sampled using the standard operating procedure (SOP) developed for product sampling events (Ecology, 2014). Products fell into five broad categories:

1. Packaging (31)
2. Paper products (26)
3. Paint and colorants (14)
4. caulks (8)
5. Miscellaneous (4) (which consisted of printer inks and food)

Products were purchased either for the packaging or the contents. For example, most of the products in the packaging category were purchased solely to sample the packaging. The contents of these products were not sampled and discarded. Caulks, however, were purchased to sample the contents. The caulk packaging was not tested.

Only three products, two mustard and one printer paper products, were sampled for both the packaging and contents. Food was not originally within the scope of this project; however, while sampling the bright yellow plastic mustard containers, the sampler decided to test the mustard contents as well. Food products are regulated by the U.S. Food and Drug Administration (FDA) and are not the responsibility of Ecology. If PCBs were found, Ecology would notify the FDA, who would have pursued the issue further.

Where possible, the printed portions of the packaging were separated from any backing paper or cardboard to concentrate the sample on the printing. The paper or cardboard backing was discarded. The packaging samples were further reduced in size using scissors and placed into sterile glass sampling jars for shipment to the laboratory. All equipment was cleaned between samples using the procedure identified in Ecology's sample preparation SOP (Ecology, 2014).

Table 3: Products tested by category

Product Packaging	Sample	Paper Products	Sample
Farfalle pasta	BARPAS	Post-It 3" by 3"	3MPOIT
Beef & Broccoli Seasoning Mix	BEFBRO	Magazine #1-cover & 1st page	BHGMAG
Berry Cereal	CAPCRU	Magazine #2-cover & first 3 pages	CONREP
Whole grain cereal	CHECAR	Magazine #3-inside pages	COOLIG
Cling wrap	CLIWRA	Business Meeting handouts	CSPAHO
Yellow mustard-bottle	FMMUTB	Multi-purpose paper	HPMUPA
Star Wars Fruit Flavored Snacks	FMSTSN	Yellow lined note pad	NOTPAD
Classic Yellow mustard-bottle	FREMUB	Local newspaper-comics	OLYCOM
Fruit by the Foot	FRUBTF	Local newspaper-front page	OLYFRO
Multi-purpose paper packaging	HPPAPA	Local newspaper-glossy inserts	OLYINS
Jello - lime	JELGRE	Local newspaper-non glossy insert	OLYNGI
Jello - lemon yellow	JELYEL	Magazine #4-back page w/yellow ad	OPRMAG
Club crackers	KEECLU	Pocket folders	POCFOL
Ancient Grains Cereal	KIANGR	Yellow report dividers	REPDIV
Sunsweet dried plums-plastic	KIRSUN	Community College Continuing Education mail	TACCCB
Sun-dried apricots	KISDAP	Tradition legal ruled yellow writing tablet-sheets	WRITAB
Lemon wafers	LEMWAF	Yellow printer paper from Ecology supplies	YELPAP
Macaroni and Cheese	MACCHE		
Milk Duds made with Chocolate & caramel	MILKDUD	Paint and Paint Pigments	Sample
Newtons Fruit Thins-plastic	NABNFT	Paint and primer	DUTBDF
Crunch granola bars	NATVAL	Interior/Exterior Spray Paint-green	HDDEGRE
Toll House Chocolate Chip-bag	NESCHO	Interior/Exterior Spray Paint-yellow	HDDEYEL
Fruit Thins	NEWTHI	Indoor/Outdoor Blue Ocean Breeze Gloss spray paint	KRYLOBL
Nilla wafers	NILWAF	Indoor/Outdoor Sun Yellow Gloss spray paint	KRYLSYE
Golden Oreos	OREOGO	Interior acrylic paint	PARPIA
Crackers Handi-Snack	RITZHS	Fluorescent neon green spray paint	RUSTFGR
No calorie sweetener	SPLEND	Fluorescent neon yellow spray paint	RUSTFYE

Natural zero calorie sweetener	STEVLE	Thalo Green colorants No. 45514Z	BEHRTGR
Taco Shells	TACSHE	TiO ₄ low VOC colorants (white) No. 46512Z	BEHRTI02
Shells & cheese packaged dinner	VELVSC	Medium yellow colorants No. 46814Z	BEHRYEL
Crackers	WHETHI	Universal paint colorant-Phthalo blue	NOVOBLU
		Universal paint colorant-Phthalo green	NOVOGRE
Caulks	Sample	Universal paint colorant-Med. Yellow	NOVOYEL
Big Stretch white	CAUBGS		
Kitchen and bath adhesive	CAUDKS	Printer Inks	Sample
Beasts the Nail Construction Adhesive	CAUDMU	Yellow ink-02XL	HP02XL
Acrylic latex plus silicone	CAUKAP	Yellow ink-564	HPYEINK
Polyseamseal all purpose	CAULOC		
White acrylic sealant plus silicone	CAURDC	Food Products	Sample
Advanced Formula Sealant	CAUSOI	Classic Yellow mustard-mustard sample	FREMUY
Phenoseal-vinyl adhesive caulk	CAUWBP	Yellow mustard-mustard sample	MEYMUU

Product samples were placed directly into glass sampling jars. For many products, this simply entailed squeezing an aliquot into the jar. For example, caulk was squeezed directly into the glass sampling jars. Spray paint was sprayed directly into glass sampling jars and allowed to accumulate until sufficient sample was available for analysis.

In addition to paint and spray paint samples, six specialty paint products (colorants) used to color paint were sampled. These colorants consisted of paint with concentrated pigment levels. Normally at the time of purchase, small aliquots of a single pigment or combination of colorants are added until the desired color is obtained. Because of their use as colorants, these specialty products would likely contain higher PCB contaminant levels than standard paint samples. Individual aliquots of the concentrated pigments, specifically yellow, green, blue, and titanium dioxide, were submitted for analysis.

Aliquots of paper samples were reduced in size using scissors and placed into glass sampling jars for shipment to the laboratory. Where possible, special emphasis was given to printed portions that contained higher amounts of yellow pigment. For example, one magazine sample consisted primarily of a yellow corporate advertisement. All equipment was cleaned between samples as identified in Ecology's sampling SOP (Ecology, 2014).

Product samples were sent to a contract laboratory for PCB analysis. Samples were analyzed using EPA Method 1668A, Chlorinated Biphenyl Congeners in Water, Soil, Sediment, Biosolids, and Tissue by HRGC/HRMS (EPA, 2010). Special care was taken to quantify all analyses for the four PCBs of primary interest (PCB-11, -206, -208 and -209) although all 209 PCB congeners were requested. This report will concentrate on the results for the four primary congeners and a separate report will be written on the full 209 congeners for all products tested.

PCB-11 Results:

Forty-nine products (66%) contained PCB-11 at detectable levels (Figure 4). The highest levels were found in packaging of a snack combining crackers and cheese (RTIZHS at 48.5 ppb), colorant added to color paint (BEHRYEL at 45.0 ppb) and yellow spray paint (HDDEYEL at 29.7 ppm). There are numerous samples in the 1 to 25 ppb and, for the 74 samples, an average concentration of 5.2 ppb was observed. More than half of the 74 samples tested contained PCB-11 above 1.66 ppb (Table 4). MRLs were consistently in the 0.1 to 1.5 ppb levels and MDLs were down to the parts per trillion (ppt) level.

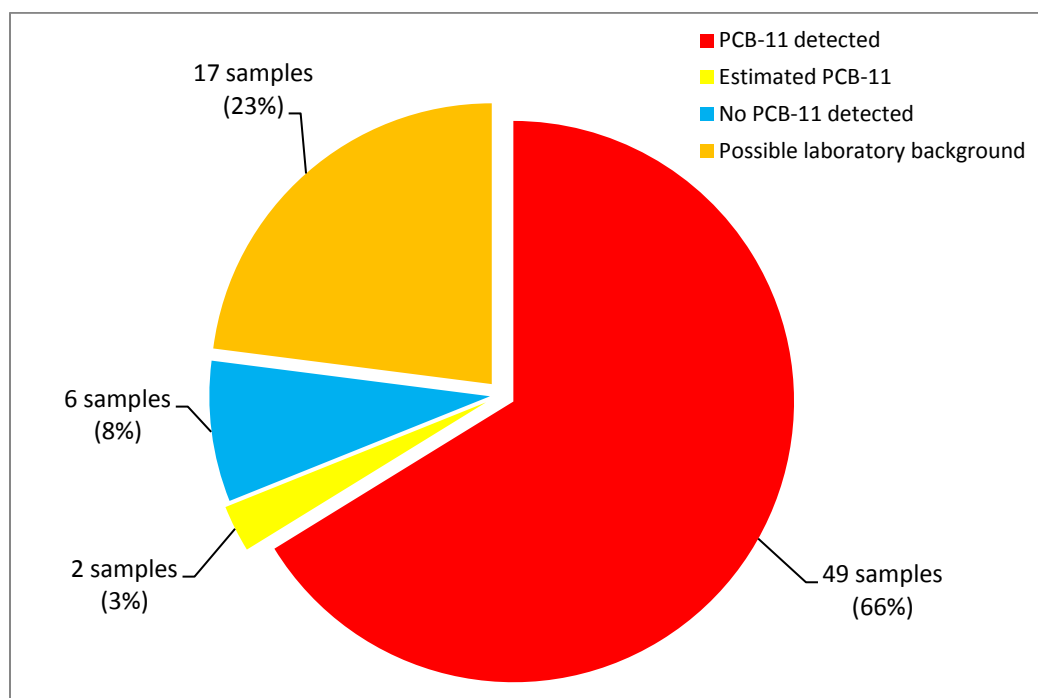
Table 4: Breakdown of PCB-11 in products

	Results	MRL	MDL
	(ppb)		
Minimum	< 0.03 ¹	0.09	0.001
Maximum	48.5	0.86	0.088
Average	5.2	0.27	0.014
Median	1.7	0.20	0.007

¹ This value reflects the MDL for this analysis. All analytical results below the MDL were assigned the MDL. For the remainder of this report, any value below the MDL is reported as 0.0.

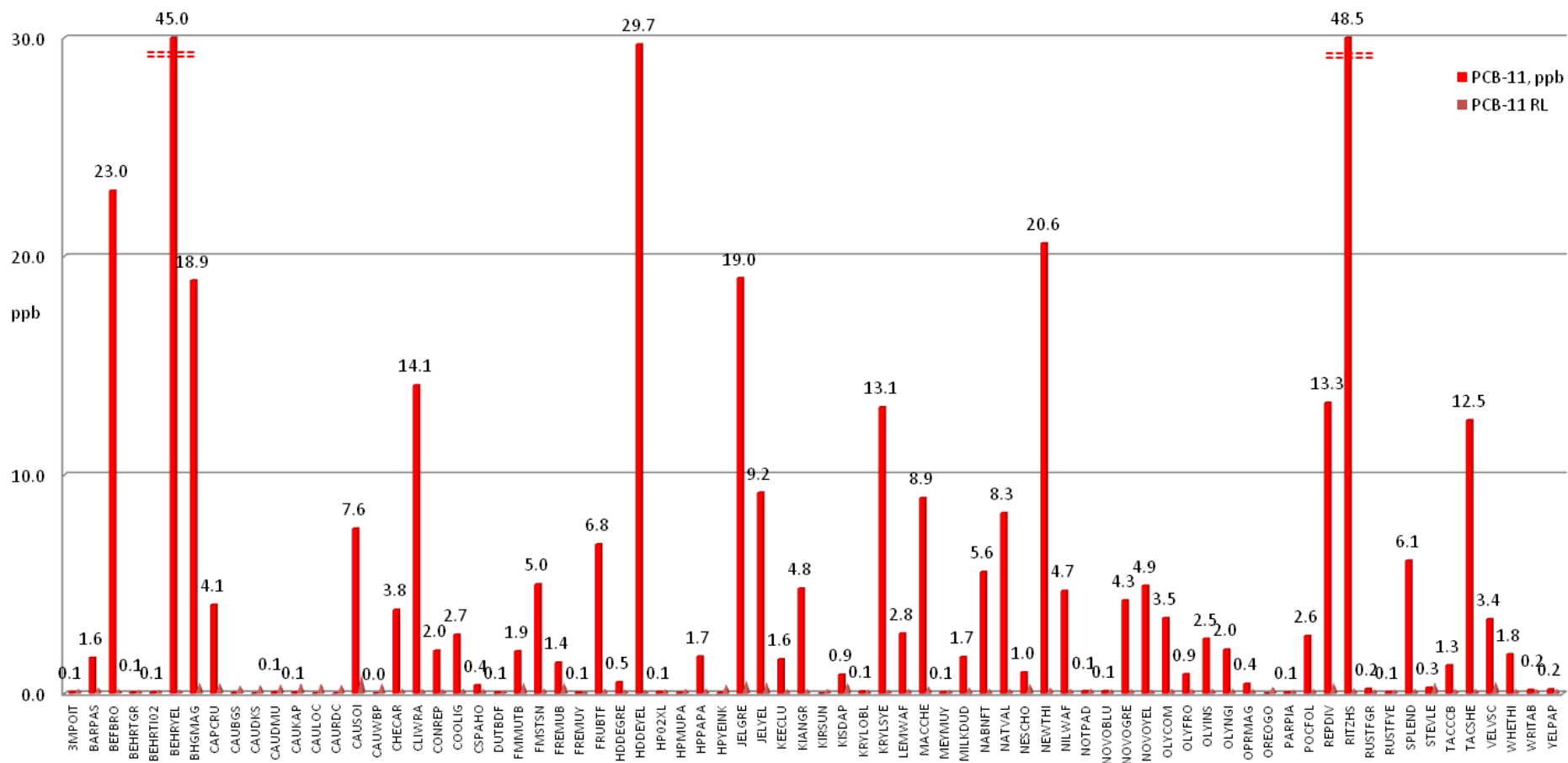
The PCB-11 results can further be separated into samples above the MRL and those between the MRL and the MDL. Any samples between the MRL and MDL are suggestive of possible presence of PCB-11 and are treated as estimates. Because of PCB-11 background levels seen in the method blanks, most of these estimated sample results, however, could be attributed to laboratory background and are subsequently disregarded. Only two of the samples, a yellow legal pad of paper (WRITAB) and packaging from a zero calorie sweetener (STEVLE), had estimated values sufficiently above the method blank results (0.1 ppb potentially attributed to laboratory background) to indicate the potential presence of PCB-11. Based upon this analysis (Figure 5), 66% of the samples contained PCB-11 above the MRL, 3% estimated values between the MDL and MRL, 8% below the MDL and 23% are likely due to laboratory background.

Figure 4: PCB-11 in consumer products



Because of broad sample variability, the products were separated into different categories which will be discussed later in this report.

Figure 5: PCB-11 concentrations in all products

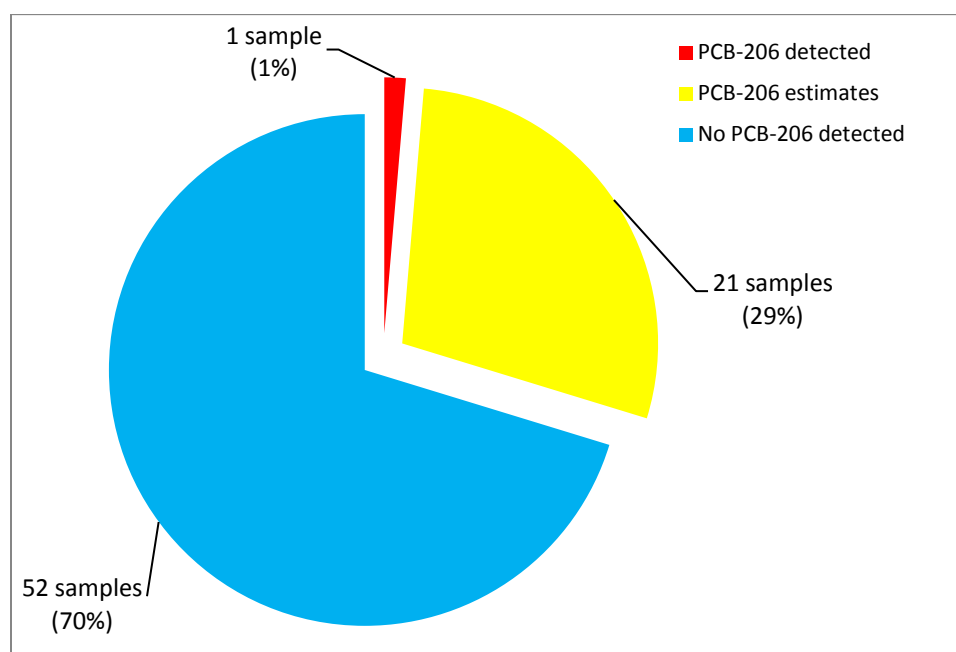


PCB-206 Results:

PCB-206 was found (Figure 6) in only one sample, a green colorant added to paint to produce a desired color, above the MRL (NOVOGRE at 5.24 ppb). Twenty-one samples (29%) reported values between the MRL and the MDL (Figure 7). Samples in this range are reported as estimates. As the method blank analyses reported no potential laboratory background issues for PCB-206, this suggests the presence of PCB-206 in these products.

Only one product, a neon yellow spray paint, contained PCB-206 near the MRL (RUSTFYE at 0.644 ppb with a MRL of 0.815 ppb). Two other samples also had PCB-206 concentrations close to the MRL, a plastic wrap container (CLIWRA) with an estimated concentration of 0.405 ppb (MRL = 0.976) and a neon green spray paint (RUSTFGR) with an estimated concentration of 0.483 ppb (MRL = 0.847). Eighteen products were near the MDL, suggestive of PCB-206 in these products. Fifty-two samples (70%) reported no detectable PCB-206 concentrations.

Figure 6: PCB-206 found in all products tested



More than half of the 74 samples tested contained PCB-206 above 0.026 ppb (Table 5), which reflects the low rate of detection for PCB-206 in the products evaluated. MRLs were consistently in 0.5 to 5 ppb levels with MDL down to the ppt levels.

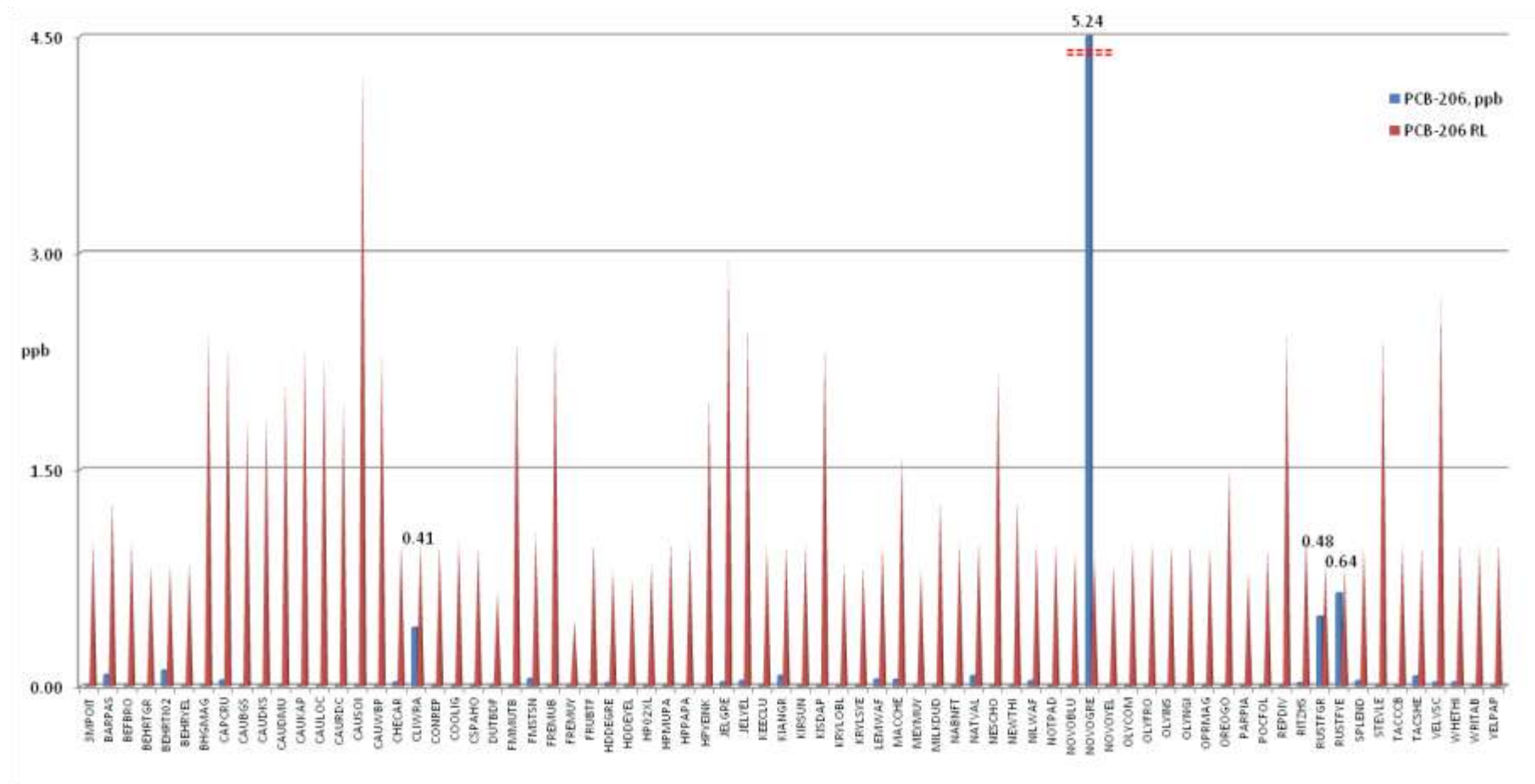
Table 5: Breakdown of PCB-206 in products

	Results	MRL	MDL
	(ppb)		
Minimum	<0.004 ²	0.456	0.004
Maximum	5.240	4.280	0.175
Average	0.122	1.358	0.024
Median	0.026	0.991	0.016

Because of broad sample variability, the products were separated into different categories, which will be discussed later in this report

² This value reflects the MDL for this analysis. All analytical results below the MDL were assigned the MDL. For the remainder of this report, any value below the MDL is reported as 0.0.

Figure 7: PCB-206 in all products



PCB-208 Results:

PCB-208 was found (Figure 9) in only one sample, green colorant added to paint to produce a desired color (NOVOGRE), at 1.41 ppb. Nineteen samples (26%) reported estimates above the MDL but below the MRL (Figure 8). As PCB-208 was found in only one of the ten method blanks (Figure 3) and this sole blank value was near the MDL (reported value of 4.7 ppt with a MDL of 3.5 ppt), these estimates are suggestive of PCB-208 presence in these samples.

Four of these 19 products were estimated to contain PCB-208 below yet near the MRL (Table 6). An additional 15 products were found near the MRL. Fifty-four samples (73%) reported no PCB-208 concentrations above the MDL.

Figure 8: PCB-208 found in all products tested

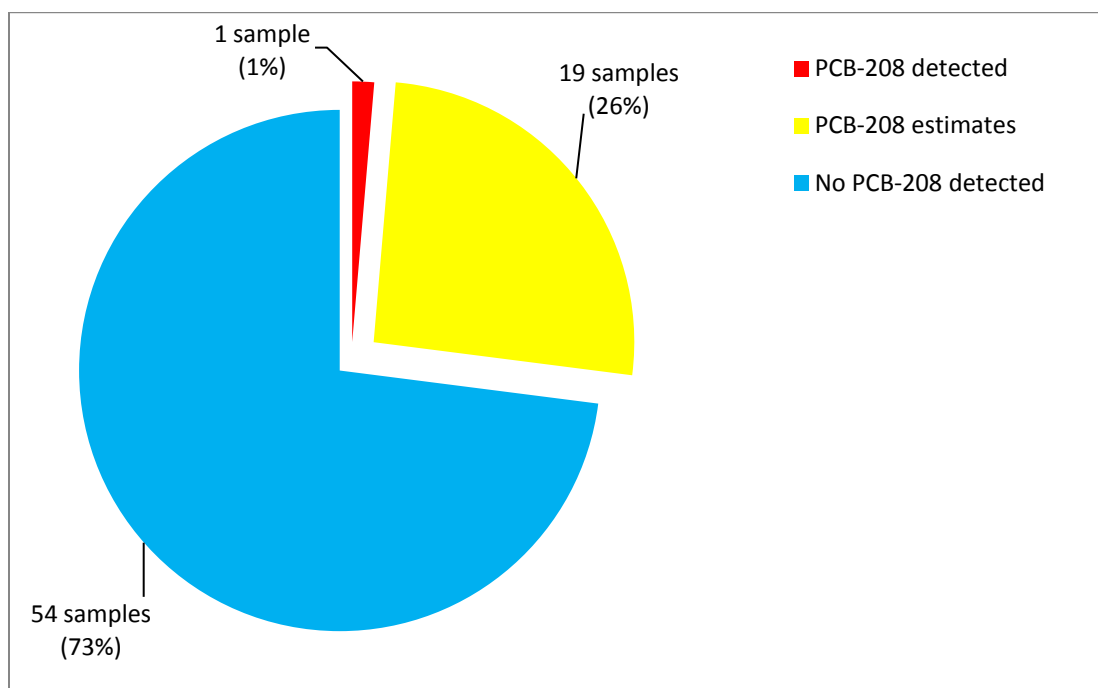


Table 6: Four products with PCB-208 estimates near the MRL

	PCB-208	Results	MRL	MDL
		(ppb)		
Neon yellow spray paint	RUSTFYE	0.605	0.815	0.015
Neon green spray paint	RUSTFGR	0.573	0.847	0.012
Beef & Broccoli Seasoning Mix packet	BEFBRO	0.600	1.000	0.033
Post-It 3" by 3"	3MPOIT	0.596	0.994	0.003

More than half of the 74 samples tested contained PCB-208 above the median 0.016 ppb (Table 7), which reflects the low rate of detection for PCB-208 in the products evaluated. MRLs were in the 0.5 to 4 ppb range with MDL down to the ppt levels.

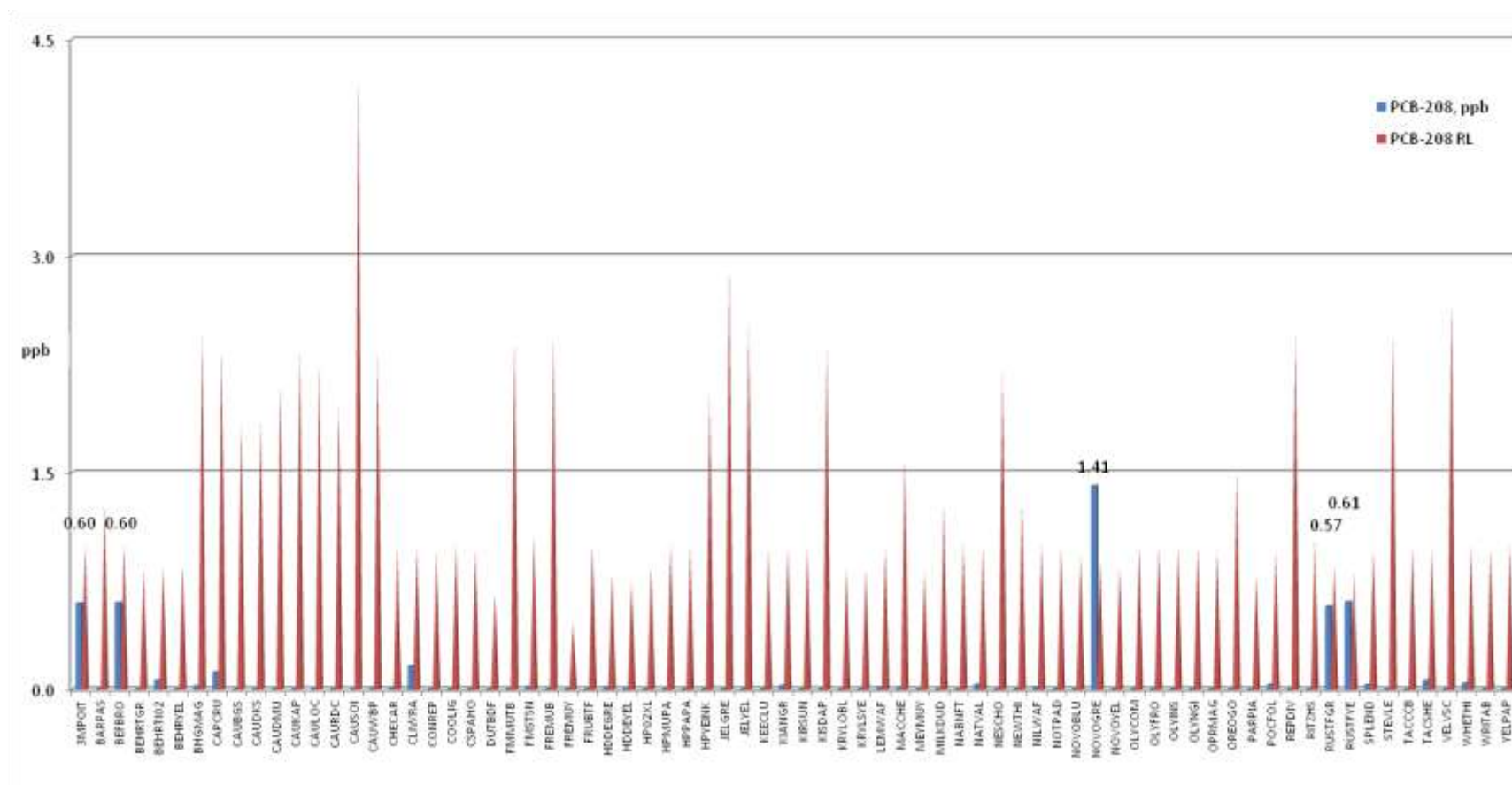
Table 7: Breakdown of PCB-208 in products

	Result	MRL	MDL
	(ppb)		
Minimum	<0.002 ³	0.456	0.002
Maximum	1.410	4.280	0.057
Average	0.070	1.358	0.013
Median	0.016	0.991	0.008

Because of broad sample variability, the products were separated into different categories, which will be discussed later in this report.

³ This value reflects the MDL for this analysis. All analytical results below the MDL were assigned the MDL. For the remainder of this report, any value below the MDL is reported as 0.0.

Figure 9: PCB-208 concentrations in all products



PCB-209 Results:

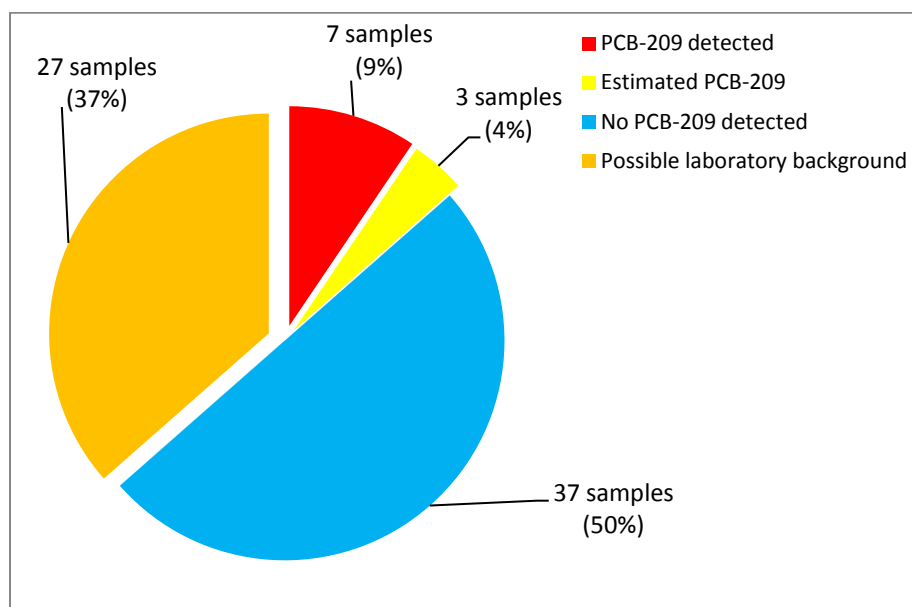
PCB-209 was found in 7 (9%) of the samples analyzed (Table 8). One sample, a green colorant added to white paint to produce a desired color, contained the highest levels of PCBs detected in all 74 samples tested with PCB-209 at 320 ppb (Figure 10). The next highest value was two orders of magnitude lower at 3.33 ppb. Six of the seven products were paint related, three spray paints and three colorants. The only non-paint related product was a yellow 2-pocket folder (POCFOL) containing PCB-209 at 1.06 ppb.

Table 8: Seven samples with PCB-209 above MRLs

Product	Name	Results	MRL	MDL
		(ppb)		
Universal colorant-Phthalo green	NOVOGRE	320.00	4.460	1.230
Fluorescent neon green spray paint	RUSTFGR	3.33	0.424	0.013
Fluorescent neon yellow spray paint	RUSTFYE	2.48	0.407	0.013
Interior/Exterior Spray Paint-green	HDDEGRE	1.27	0.402	0.008
TiO ₂ low VOC colorant No. 46512Z	BEHRTI02	1.26	0.421	0.021
2-pocket folders	POCFOL	1.06	0.477	0.006
Thalo Green colorant No. 45514Z	BEHRTGR	1.00	0.417	0.016

Thirty samples were reported as estimates. Twenty-seven samples (37%) were below the 0.1 ppb (Figure 11), indicative of potential laboratory background and cannot be taken as suggestive of PCB-209 presence. It is unlikely the remaining three estimates were due to laboratory background. The highest value was for a green cracker box (KEECLU) at 0.207 ppb (MDL = 0.485). The other two estimates included a paint (DUTBDF) and printer ink (HP02XL) at 0.184 and 0.177 ppb, respectively. These estimates are suggestive of PCB-209 presence in these samples. Thirty-seven samples (50%) reported no detectable levels of PCB-209.

Figure 10: PCB-209 found in all products tested



More than half of the 74 samples tested contained PCB-209 above 0.018 ppb (Table 9), which reflects the low rate of detection for PCB-209 in the products evaluated. MRL were in 0.2 to 4.5 ppb levels with MDL down to the ppt levels.

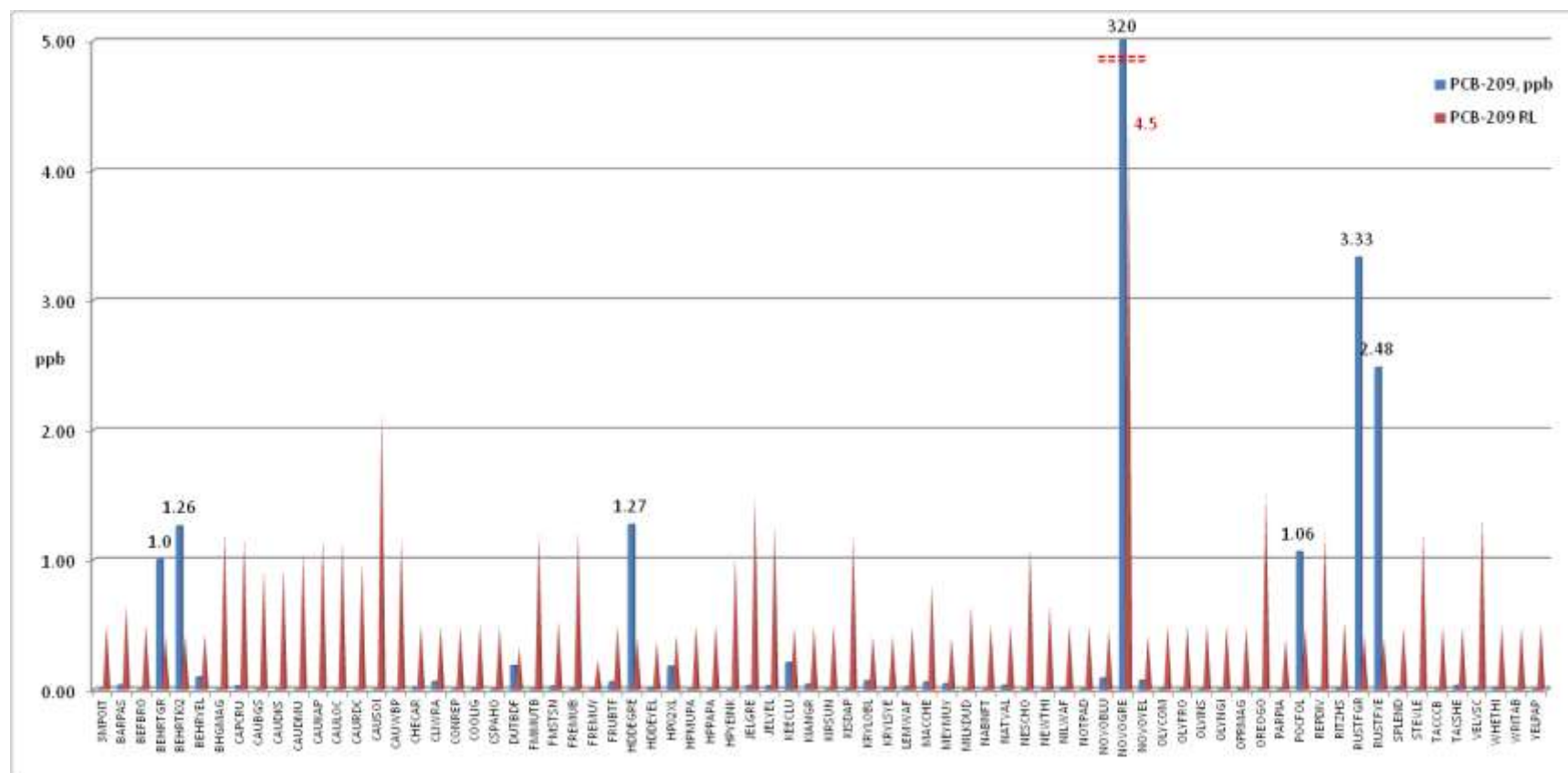
Table 9: Breakdown of PCB-209 in products

	Result	MRL	MDL
	(ppb)		
Minimum	<0.002 ⁴	0.228	0.002
Maximum	320.000	4.460	1.230
Average	4.492	0.744	0.029
Median	0.018	0.496	0.008

Because of broad sample variability, the products were separated into different categories, which will be discussed later in this report

⁴ This value reflects the MDL for this analysis. All analytical results below the MDL were assigned the MDL. For the remainder of this report, any value below the MDL is reported as 0.0.

Figure 11: PCB-209 in all products

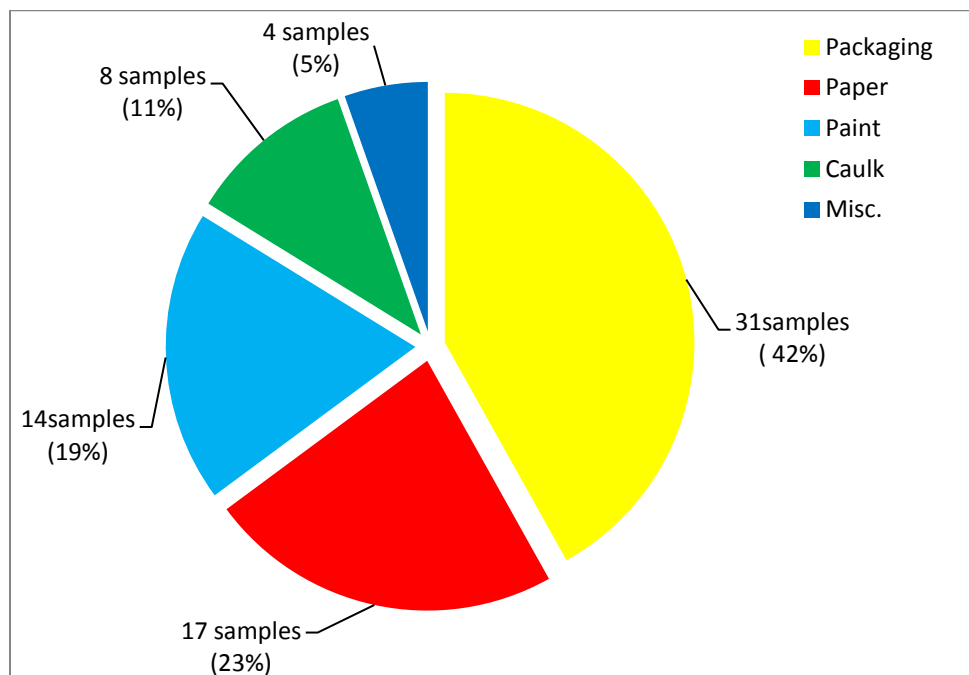


Results by Product Category

For this report, the analytical results are separated into five product categories (Figure 12):

1. Product packaging (31 products)
2. Paper products (17)
3. Paint and paint products (14)
4. Caulks (8)
5. Miscellaneous-food products (2) and printer inks (2)

Figure 12: Sixty-eight product types



Product packaging includes both paper and plastic bottles and wrapping used to protect products. Paper products include newspaper and office supplies. All paints and painting products are grouped together regardless of color. Initially, the products were separated into six categories related to the primary color involved. Categories included yellow, blue, green, white, and yellow with substantial other colors and multiple colors.

The groups were evaluated for variability and PCB distribution. No conclusions could be drawn.

- PCB-11, for example, appeared in all product categories and is likely the result of a multi-step color process that includes PCB-contaminated pigments and dyes in more than one step.
- There were insufficient blue and green products to reach any firm conclusions.
- The caulks are grouped with the white.
- Food was not within the original scope of this project; however, while sampling the plastic containers, the sampler decided to submit the bright yellow product (mustards) for analysis to determine if mustard also contained PCB congeners. Each grouping is discussed separately.

Product Packaging

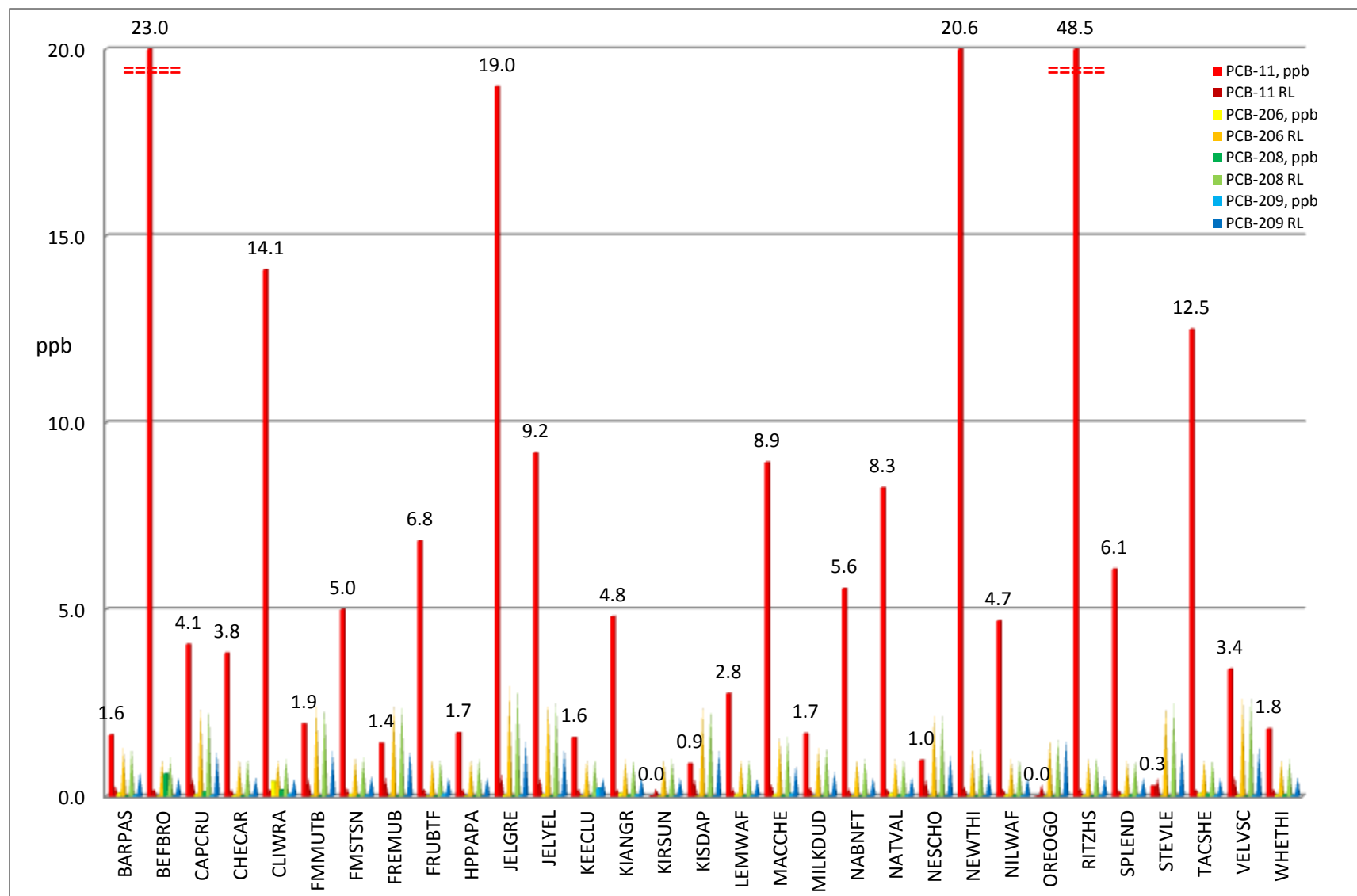
The 31 product packaging results are shown in Figure 13⁵. PCB-11 was found in all but two products. One product, a cracker and cheese snack pack (RITZHS), contained the highest concentrations of PCB-11 found in this study at 48.5 ppb. There were several products with appreciable PCB-11 concentrations and the average PCB-11 concentration for this category was 7.26 ppb. Only one sample contained an estimated PCB-11 of 0.27 ppb.

No product packaging samples contained detectable levels of PCB-206, -208, and -209. There were a few estimated values, which suggests the possible presence of these congeners in product packaging, but because of the limited sample size, additional sampling is needed.

Lastly, the dominant color of the package may not be predictor of congener distribution. A primarily green (KEECLU) and red (KIANGR) packaging both contained substantial amounts of PCB-11 at 1.57 and 4.81 ppb, respectively. This suggests either that many pigments contain PCBs or the four-color printing process includes sufficient amounts of PCB-contaminated pigments to impact any product packaging. The printing process may be more of a factor than the specific pigments used, although no information was available on what pigments were used for these specific products.

⁵ Note: The product sampling results are shown as columns and the MRLs shown as spikes. There are different though related color differences as well. PCB-11 is bright red while the PCB-11 MRL is dark red.

Figure 13: PCBs in product packaging



Paper Products

The PCB results for 17 paper products are shown in Figure 14⁶. PCB-11 was found in all products either as values or estimates. However, three products (Table 10) reported estimated values that, because of potential issues identified with the sample blanks, may be the result of laboratory background and not due to the presence of PCB-11. Two additional products, a yellow legal pad (WRITAB) and yellow printer paper (YELPAP), reported estimated values very close the MRL (Table 10). These products suggest the presence of PCB-11 albeit at low levels, which might still impact water quality discharge limits. Two products, Magazine #1 (BHGMAG) and yellow report dividers (REPDIV) contained PCB-11 at levels of 18.9 and 13.3 ppb, respectively. The average PCB-11 concentration for these products was 3.01 ppb.

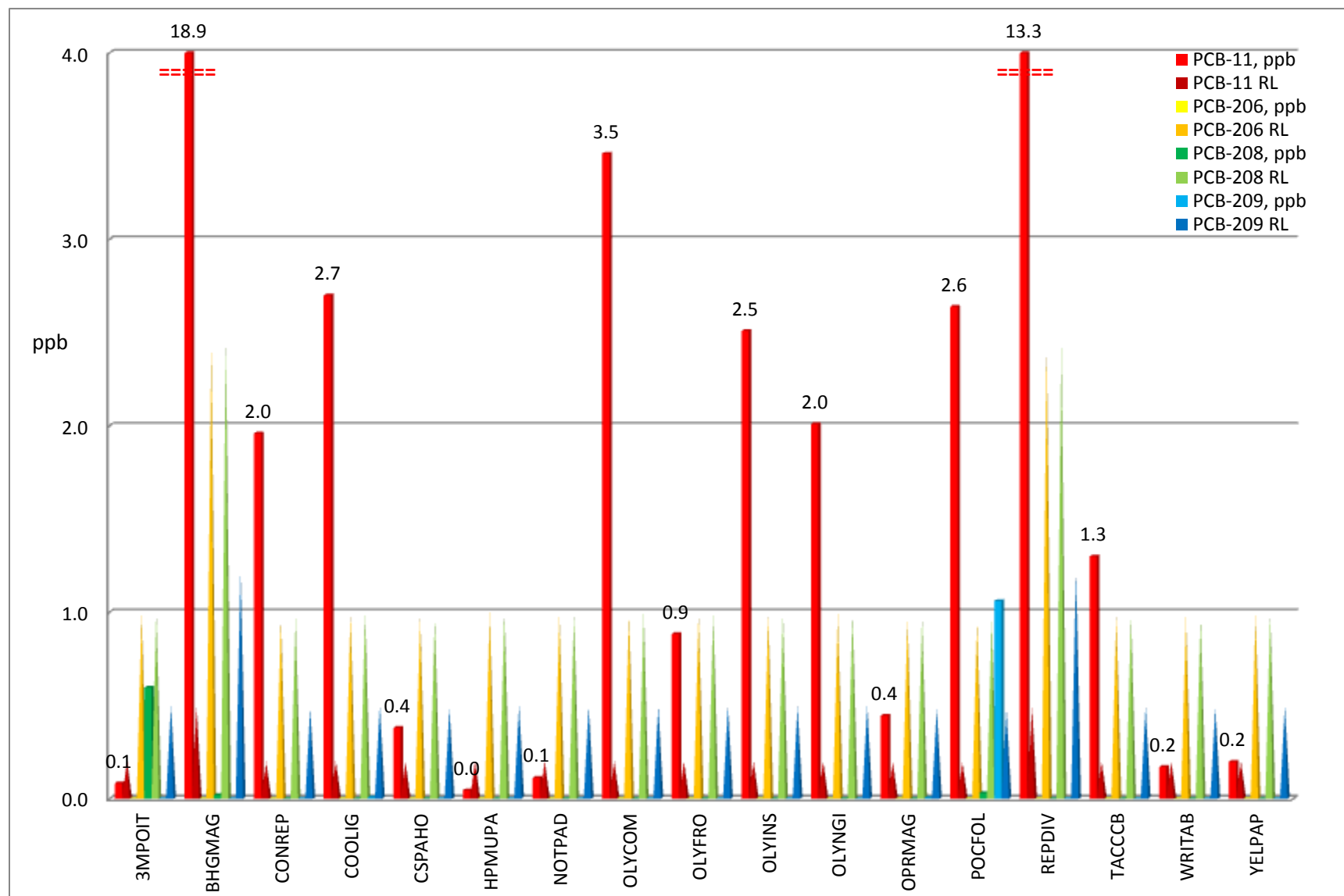
Table 10: Three products potentially affected by laboratory background or near MRL

Product	Name	Results	MRL	MDL
		(ppb)		
Potential Laboratory background				
Post-It 3" by 3"	3MPOIT	0.08	0.20	0.003
Multi-purpose white paper	HPMUPA	0.05	0.20	0.010
Yellow lined note pad	NOTPAD	0.11	0.20	0.009
Products Near MRL				
Tradition legal yellow writing tablet	WRITAB	0.17	0.19	0.006
Yellow printer paper	YELPAP	0.20	0.20	0.013

No samples contained either PCB-206 or PCB-208 above the MRL, although three samples contained estimated PCB-208 values. Only one of these samples, the small Post-It (3MPOIT), reported a value substantially above the MDL at 0.60 ppb (MRL=0.99). The other two samples were near the MDL and can be assumed to be indicative of potential PCB presence. One sample, a yellow pocket folder, did contain PCB-209 above the MRL at 1.06 ppb (MRL=0.48). Two other samples reported estimated values of PCB-209; however, because of potential laboratory background issues, these results may be caused by laboratory background and may not be an indication of PCB-209 in these products. Because of the limited sample size, additional sampling is needed.

⁶ Note: The product sampling results are shown as columns and the MRLs shown as spikes. There are different though related color differences as well. PCB-11 is bright red while the PCB-11 MRL is dark red.

Figure 14: PCBs in paper products



Paint and paint products

The PCB results for 14 paints and colorants are shown in Figure 15⁷. PCB-11 was detected in 7 of the 14 products (50%). Four products are yellow and three are green. Five of those 7 (36%) contained appreciable concentrations ranging from 4.3 to 45.0 ppb while the remaining two were found above the MRL but below 1 ppb. Four of the yellow samples found at appreciable levels were either yellow paint used to color white paint (BEHRYEL, HDDEYEL, and NOVOYEL) or a neon yellow spray paint (KRYLSYE). One yellow colorant (BEHRYEL) reported the second largest PCB-11 concentration observed in this study at 45.0 ppb.

The only non-yellow paint product to exhibit appreciable levels of PCB-11 was a phthalo green colorant (NOVOGRE) at 4.3 ppb. As this was the only product to report all four PCB congeners at appreciable concentrations, it will be discussed more later in this section. Seven products contained estimated values; however, because of sample blank issues described previously, six of them may have resulted from laboratory background during analysis and cannot be taken as a possible indication of PCB-11 presence. Only a neon green spray paint (RUSTFGR) reported an estimated value at 0.113 ppm (MRL=0.18), sufficiently near the MRL to suggest the possible presence of PCB-209. The average PCB-11 concentration for these products was 7.02 ppb.

Only one product, the green colorant added to color white paint (NOVOGRE) contained detectable levels of PCB-206. This product will be discussed more later in this section. Four other products contained estimated PCB-206 concentrations. PCB-206 was not found in any of the sample blanks so there is no expected laboratory background. One estimate, a green spray paint (HDDEGRE), reported PCB-206 at 0.02 ppb, only slightly above the MDL (0.015 ppb). This sample may not be a true indication of the presence of PCB-206. The other three, all spray paints (BERHTI02, RUSTFGR, and RUSTFYE) detected estimated values sufficiently above the MDLs to suggest the presence of PCB-206.

Similar results are reported for PCB-208 as for PCB-206. The same paints contain the same relative amounts of PCB-208. The only major difference is that the white paint colorant (BEHRTI02) contains only about half the amount of PCB-208 at 0.06 ppb (MDL=0.044). This concentration is close to the MDL, so it cannot be assumed this product contains PCB-208.

The results for PCB-209 are noticeably different. Thirteen of the 14 products contain some level of PCB-209, seven of which are qualified as estimates. Of the products containing PCB-209, one, the green colorant used to color white paint (NOVOGRE), has the highest concentration of any PCB in this study at 320 ppb (MRL = 4.46). This product is discussed in more detail later in this section. The remaining five products all have PCB-209 concentrations ranging from 1.0 to 3.3 ppb and include two more paint colorants added to color white paint (BEHRTGR and BEHRTI02) and three spray paints (HDDEGRE, RUSTFGR and RUSTFYE). The remaining seven products contained estimated concentrations. Six have PCB-209 levels that, because of laboratory background issues, may not indicate the presence of PCB-209. Only one paint sample (DUTBDF) has a concentration of 0.184 ppb (MRL=0.327 ppb) to

⁷ Note: The product sampling results are shown as columns and the MRLs shown as spikes. There are different though related color differences as well. PCB-11 is bright red while the PCB-11 MRL is dark red.

suggest the presence of PCB-209. Only one sample, a white interior latex paint (PARPIA) contained no reported or estimated concentrations of any of the four congeners of interest.

The phthalo green colorant used to color white paint (NOVOGRE) contained all four PCBs above MDL levels (Figure 16) and was unique in this study. Of the four PCBs, only PCB-208 was near the MDL while the other three were appreciably above the MRL. The reason for this distribution is unknown; however, based upon these results, it suggests that this particular pigment is highly contaminated with PCBs. More research into the pigment is warranted and future studies may direct more sampling towards products containing this pigment.

Figure 15: Four PCB congeners detected in a phthalo-green colorant The columns represent PCB concentrations and the spikes the corresponding MRL.

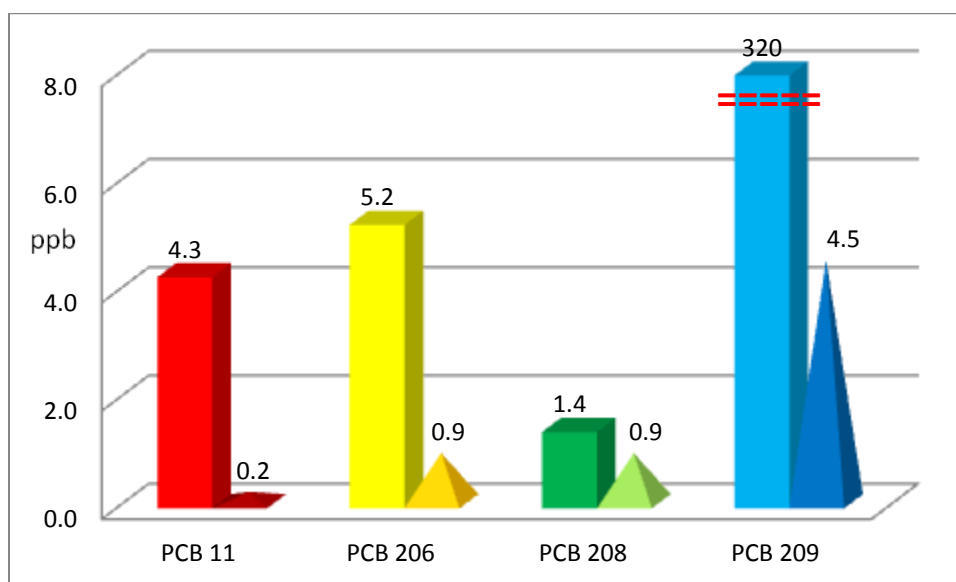
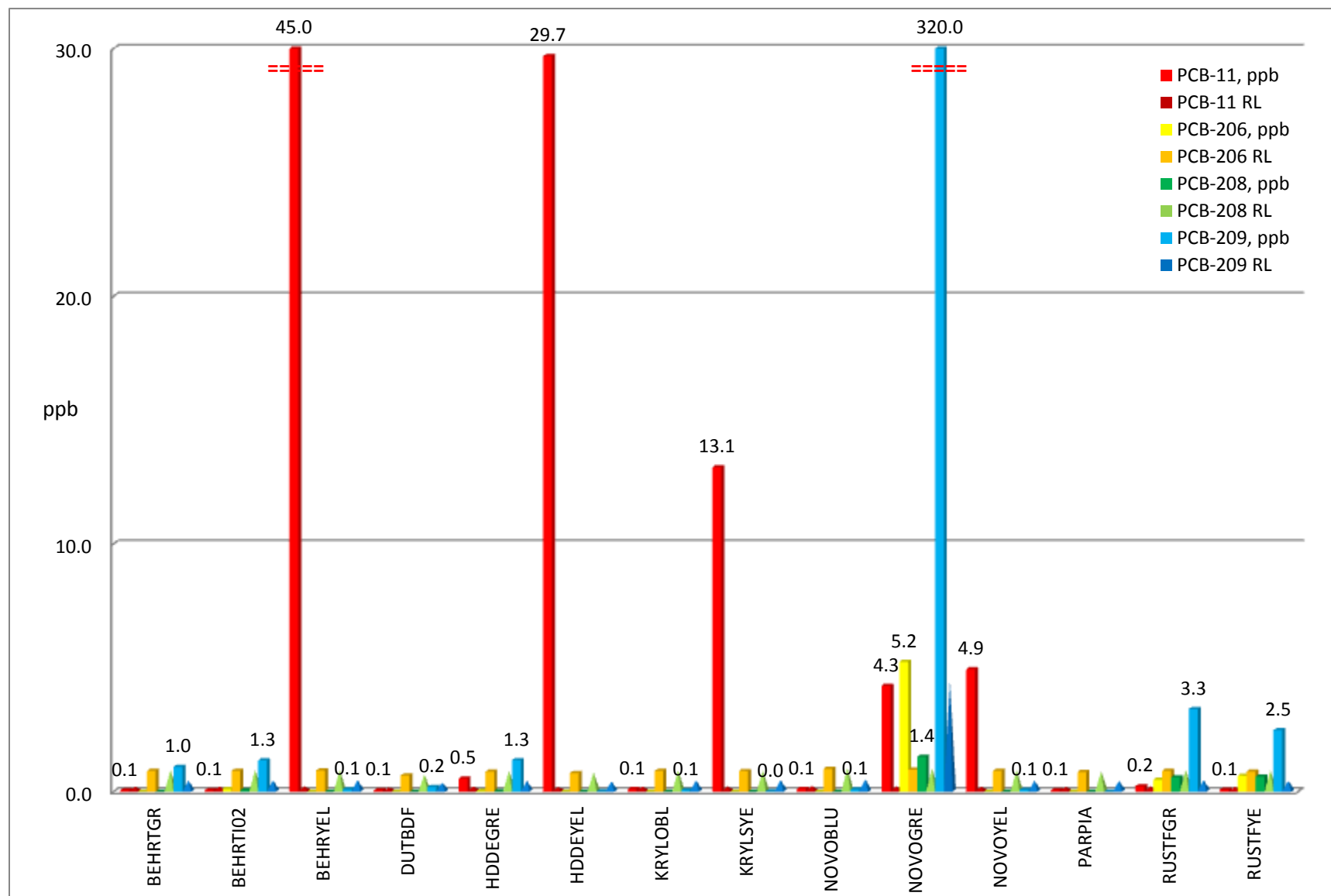


Figure 16: PCBs in paint and paint products



Caulks

Eight caulks were purchased from a large national home improvement store and sampled (Table 11). The caulks were selected to represent the variety of products available at a specific store but are only a fraction of the caulks available. Many of the caulks contained cautions against coming in contact with eyes, skin, inhaling or swallowing. Many indicated they were skin and respiratory irritants. Two of the caulks were colored. CAURDC transformed from pink to white as it cured and CAUWBP was colored beige.

Table 11: Caulk types selected for analysis

Sample ID	Description
CAUBGS	Stretching caulk-seals doors, windows and siding
CAUDKS	Kitchen and Bath adhesive caulk-provides waterproof seal
CAUDMU	Construction adhesive-general construction, remodeling, maintenance and repair
CAUKAP	Acrylic latex caulk plus silicon-indoor/outdoor use
CAULOC	Polyseamseal all purpose indoor/outdoor adhesive-seals, bonds and fills
CAURDC	ColorCure Pink 2 White acrylic sealant plus silicone-all purpose
CAUSOI	Advanced formula sealant-window, door and siding
CAUWBP	Vinyl adhesive caulk-indoor/outdoor use 'does it all'

The PCB results for caulks are shown in Figure 17. As indicated previously, only new caulks were sampled and these products are indicative of caulks currently for sale and use. PCB-11 was found in only one product (CAUSOI) at a level of 7.6 ppb. The remaining samples either contained no PCB-11, or in three cases, reported estimates just above the MDL ranging from 0.03 to 0.06 ppb. Because of potential laboratory background issues, these three results are not an indication of possible PCB-11 presence in these samples. None of these samples reported the presence of PCB-206, -208, or -209.

CAUSOI is the only caulk tested that describes itself as being pink when applied but cured to white when dry. The pink coloring possibly accounts for the presence of PCB-11. No PCB-11, however, was found in the beige-colored caulk. Because of the small sample size, further sampling is needed to determine whether caulks and particularly colored or color-changing caulks contain PCBs of interest.

Miscellaneous

The PCB results for printer inks and food are shown in Figure 18. As indicated previously, these products are in a miscellaneous category because the sample cohort is too small to reach any definitive conclusions. In addition, food samples were not part of the original scope of this study but analyzed as a sample of opportunity. Food contents are regulated by the U.S. Food and Drug Administration and are outside of Ecology's area of concern.

All four products have estimated levels of PCB-11 and PCB-209. With the exception of one sample, these concentrations are possibly due to laboratory background issues and may not indicate the presence of PCBs in these products. The only exception is PCB-209 in one of the inks (HP02XL) with a concentration of PCB-209 at 0.18 ppb (MDL = 0.025). This value is sufficiently above the MDL laboratory background to suggest the presence of PCB-209. Further study is warranted to confirm the presence of PCB-209 in this type of sample.

Figure 17: PCBs in caulks

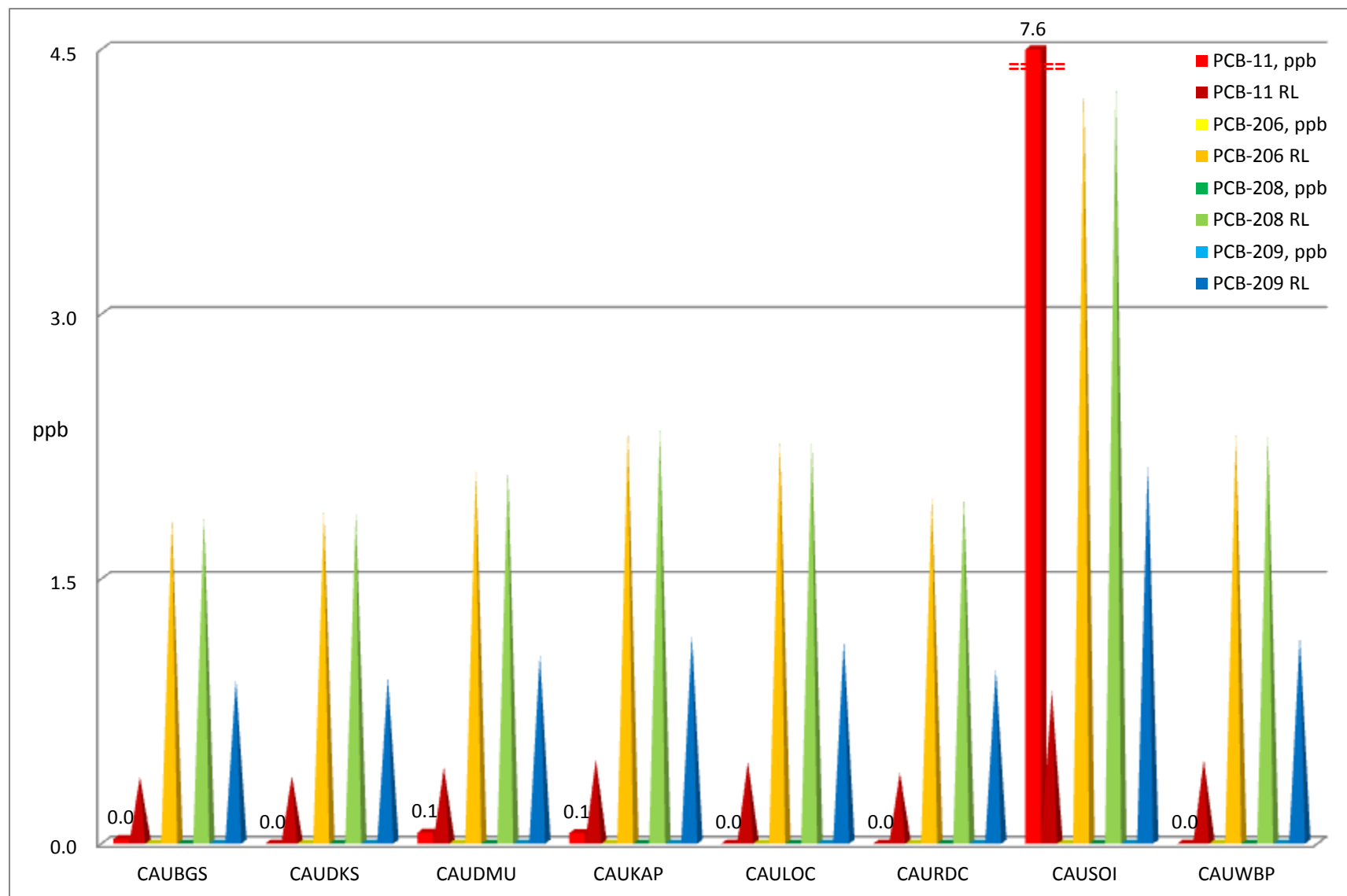
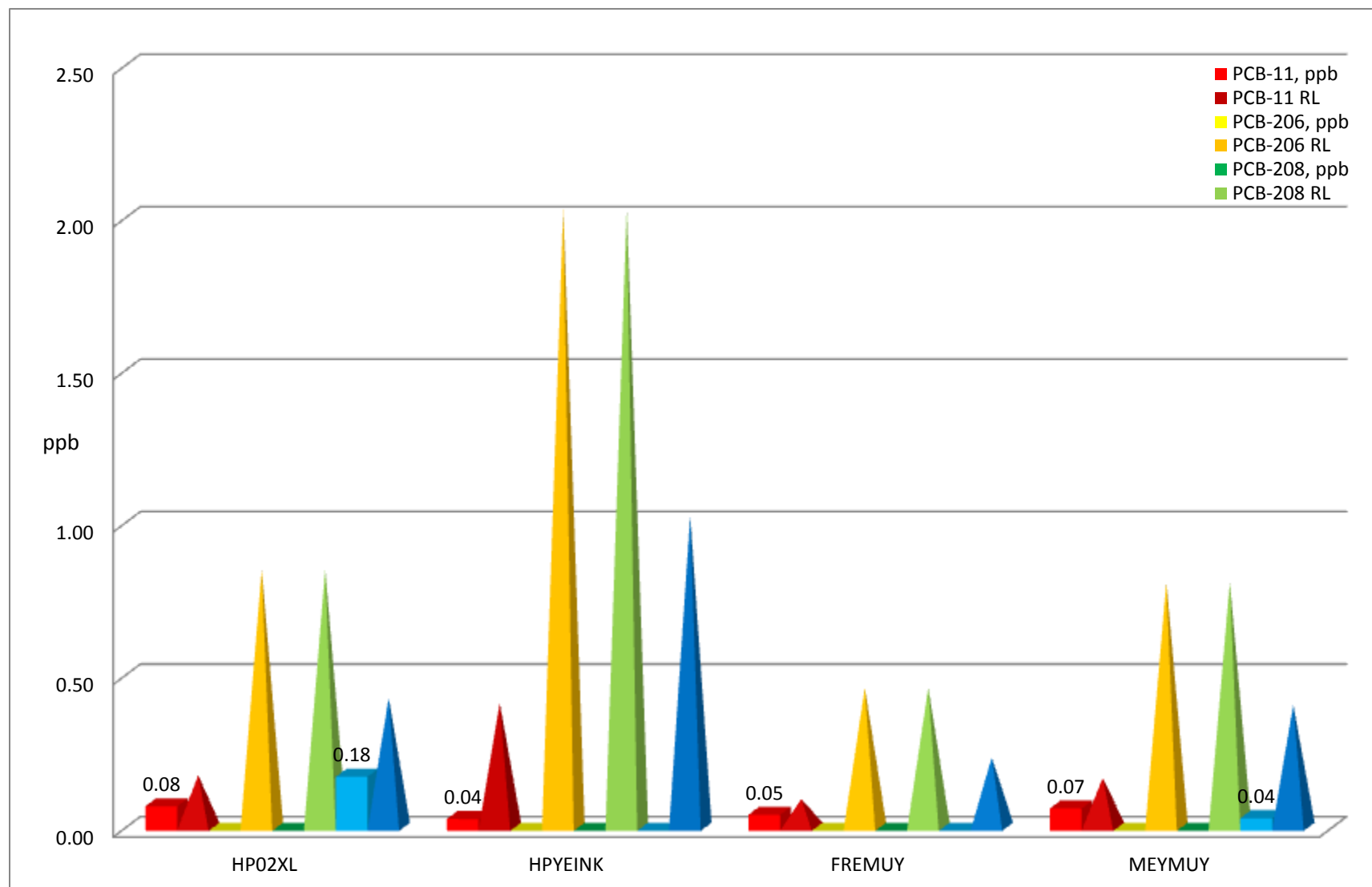


Figure 18: PCBs in miscellaneous products (printer inks and food)



Conclusions

Based on the results described above, the following conclusions can be reached.

- PCB-11 is found in a wide range of product types and at measurable concentrations. This includes product packaging, paper products, and paint and colorants. Many of the products contain PCB-11 in the 5 to 45 ppb levels.
- Given the wide distribution of PCB-11 in consumer products, consumer products are a continuing and new source of PCB contamination and generation of PCB-11 is mostly an unregulated source of PCB contamination.
- Some green and blue colorants used to color paint contain appreciable concentrations of PCB-209. One phthalo green colorant contained all four PCB congeners at detectable concentrations and reported the highest level of any product tested with a detectable level of PCB-209 at 320 ppb.
- PCB-206 and -208 were not detected in most products. One colorant contained detectable levels of all four PCBs. This product accounts for the only PCB-206 and -208 detectable in the consumer products analyzed.
- Based upon a very limited sample size, white caulks do not appear to contain the four PCBs of interest. Based upon a single sample, caulks that change color during curing may be a source of PCB-11. Additional sampling is needed to better define the potential PCB contamination found in caulks.
- Areas for future study include yellow, green, and blue paints and colorants, colored clothing, cosmetics, soaps and hand sanitizers, and household cleaning products.

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Appendix A

Acronyms and abbreviations used in this report

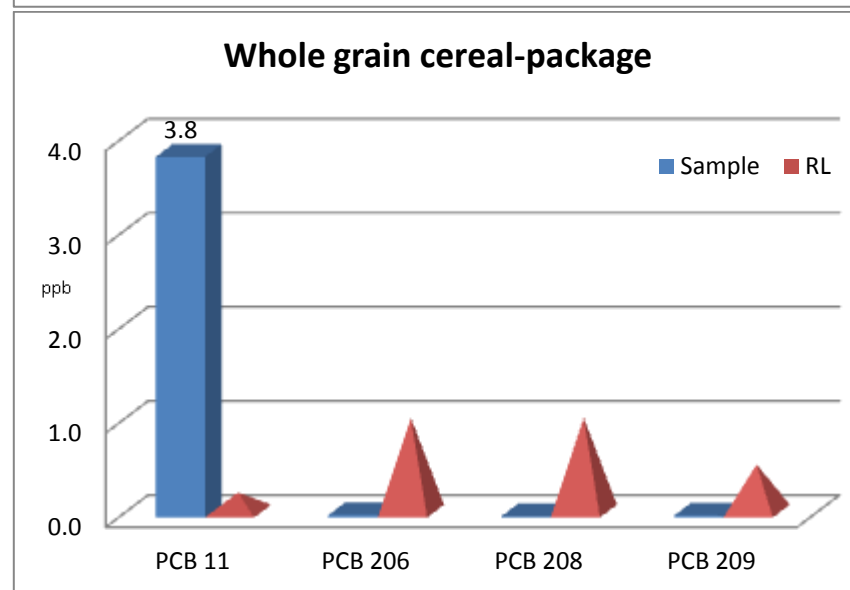
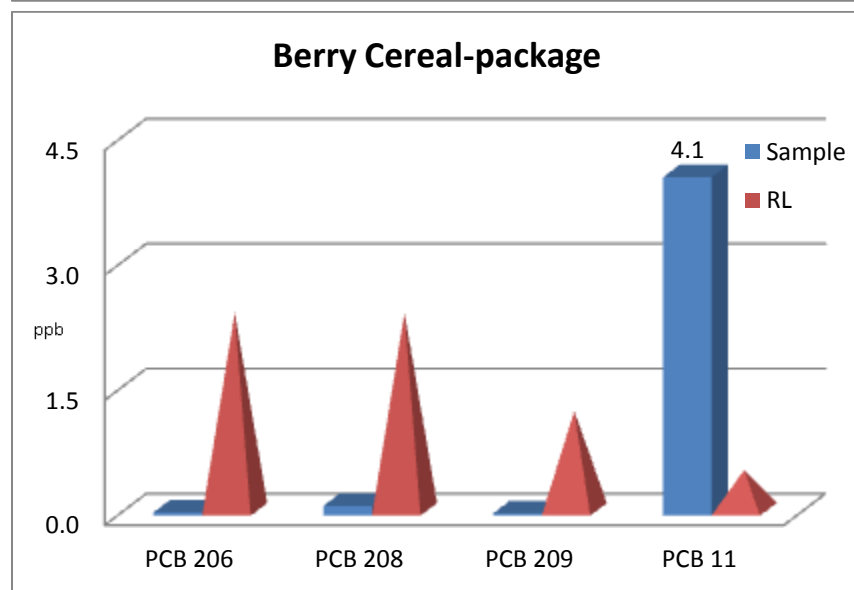
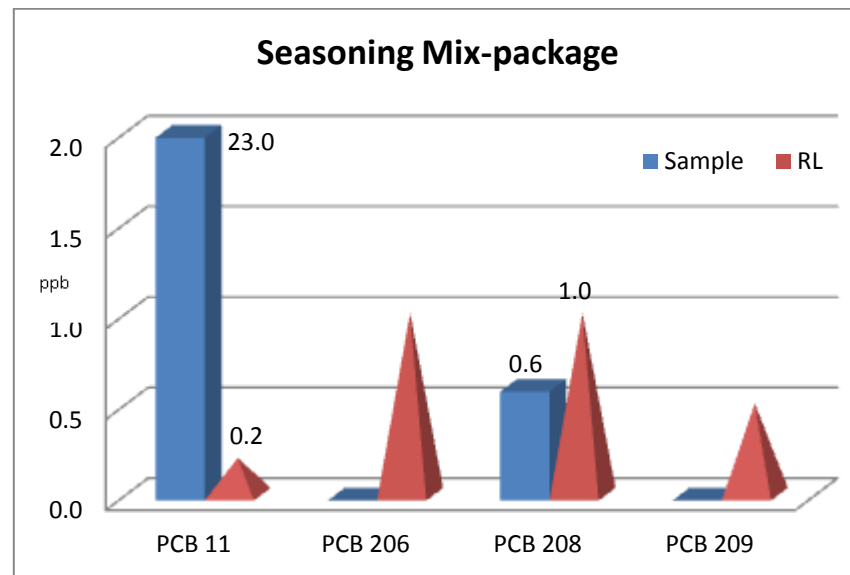
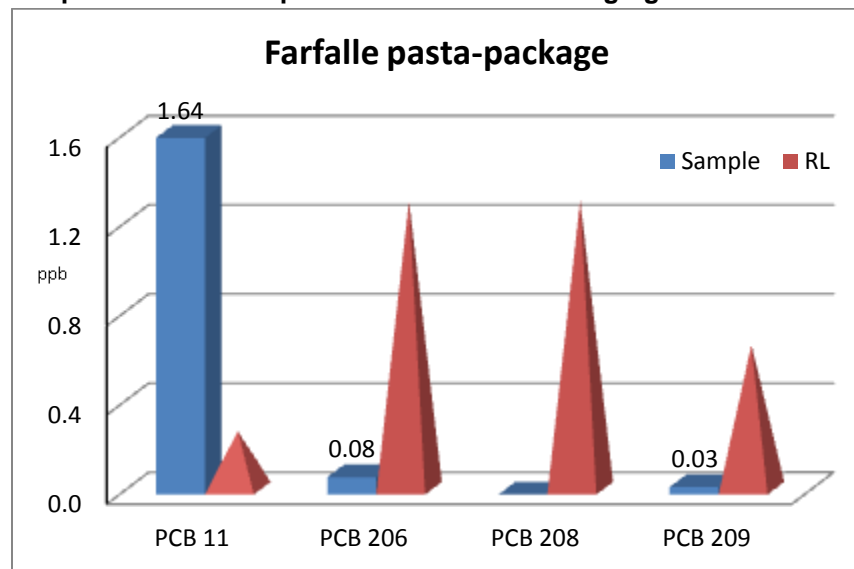
DOH	Washington State Department of Health
EAP	Environmental Assessment Program
EC	Environment Canada
Ecology	Washington State Department of Ecology
EPA	United States Environmental Protection Agency
et al.	Et alia or and others
EU	European Union
GC-MS	Gas Chromatography-Mass Spectroscopy
HCl	Hydrochloric acid
HF	Hydrofluoric acid
HNO ₃	Nitric acid
HQ	Headquarters
HWTR	Hazardous Waste and Toxics Reduction Program
i. e.	Id est or In other words
LCS	Laboratory control sample
LOQ	Limit of Quantitation
MEL	Manchester Environmental Laboratory
MDL	Method detection limit
MQO	Measurement quality objective
MRL	Method reporting limit
NEP	National Estuary Program
NICNAS	National Industrial Chemicals Notification and Assessment Scheme
OECD	Organisation for Economic Cooperation and Development
PCB	Polychlorinated biphenyls
PBT	persistent, bioaccumulative, and toxic substance
PPB	Parts per billion
PPM	Parts per million
PQL	Practical quantitation limit
RCW	Revised Code of Washington
RDP	Resorcinol diphenyl phosphate
RL	Reporting limit
QA	Quality assurance
QC	Quality control
QAPP	Quality Assurance Project Plan
RPD	Relative percent difference
RSD	Relative standard deviation
SOP	Standard operating procedures
SRM	Standard reference materials

Units of Measurement

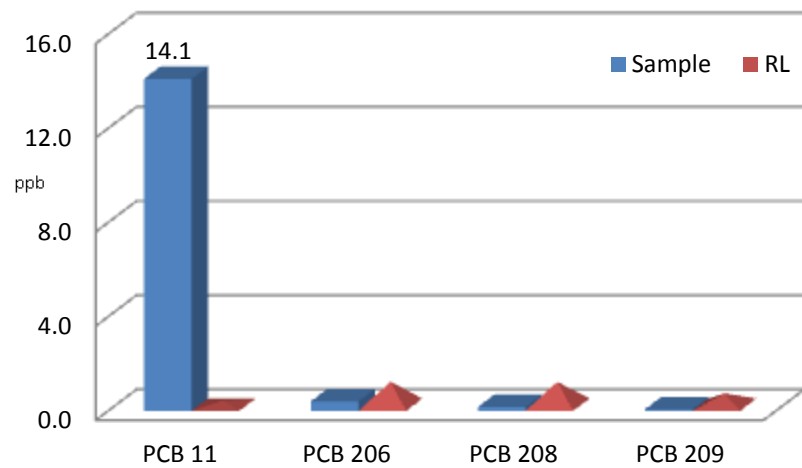
ng	nanogram, a unit of mass equal to one millionth of a gram
mg	milligram, one thousandth of a gram
g	gram, a unit of mass
kg	kilograms, a unit of mass equal to 1,000 grams.
meter	meter, a unit of distance
mm	millimeter, a unit of distance equal to one thousandth of a meter
Liter	liter, a unit of volume
mL	milliliter, equal to one thousandth of a liter
ppb	parts per billion
ppm	parts per million
mg/kg	milligrams per kilogram (parts per million)
ng/g	nanograms per gram (parts per billion)
ng/kg	nanograms per kilogram (parts per trillion)
mg/L	milligrams per Liter (parts per million)
ng/L	nanograms per Liter (parts per trillion)
s.u.	standard units

Appendix B

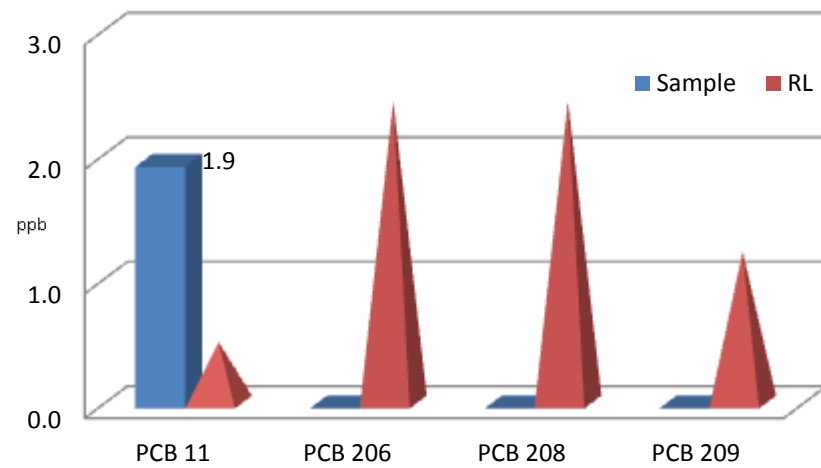
Graphs of individual products - Product Packaging



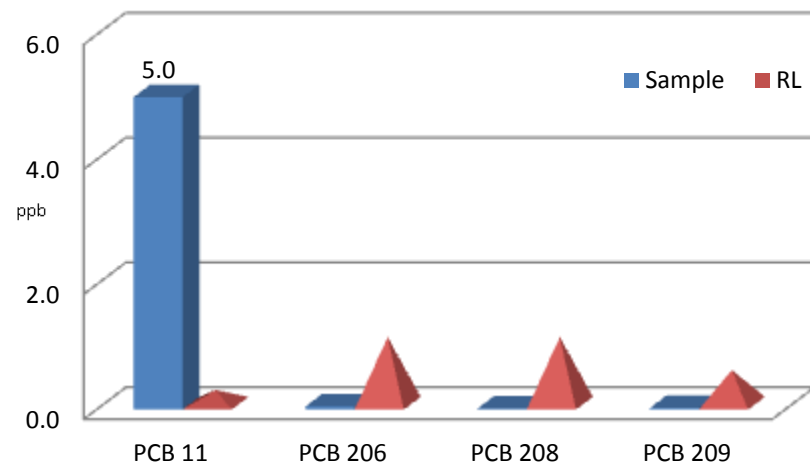
Cling wrap-package



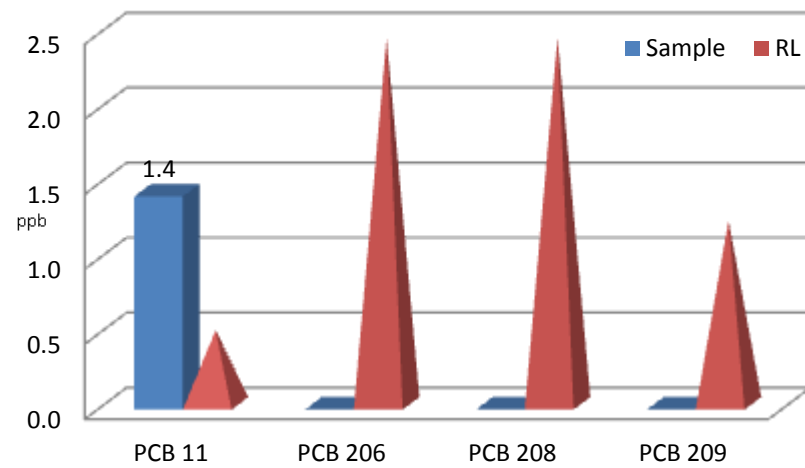
Yellow mustard-bottle #1



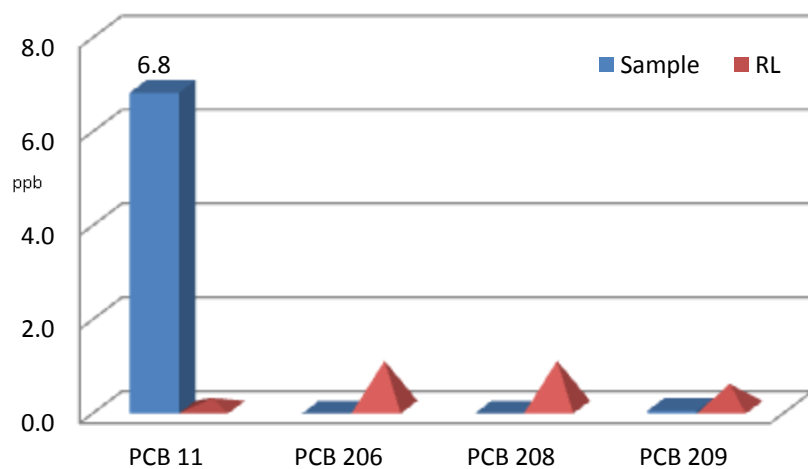
Fruit Flavored Snacks-package



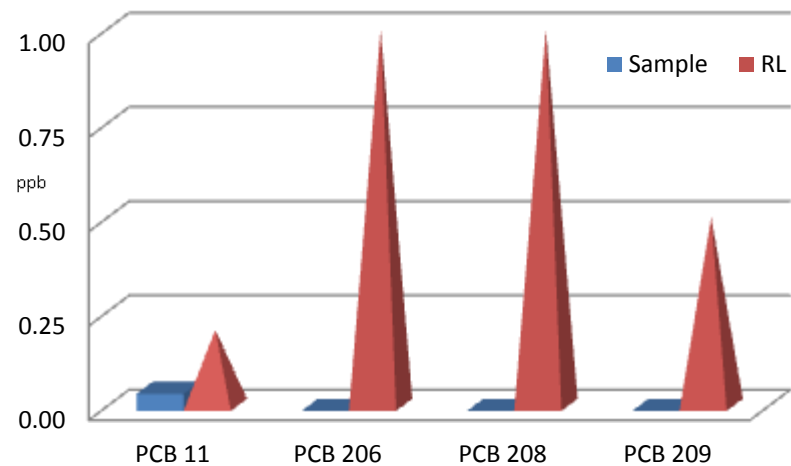
Yellow mustard-bottle #2



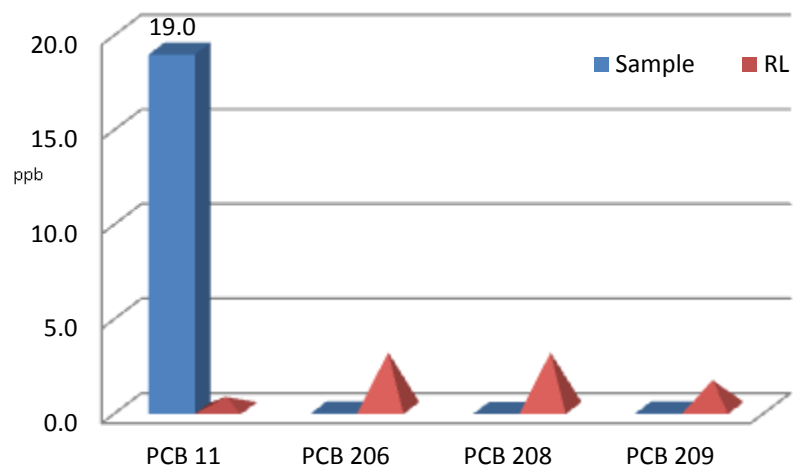
Fruit by the Foot-package



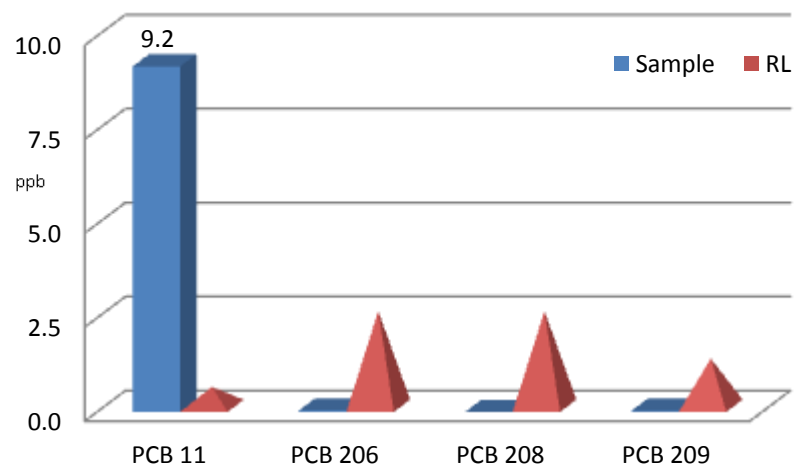
Multi-purpose paper-package



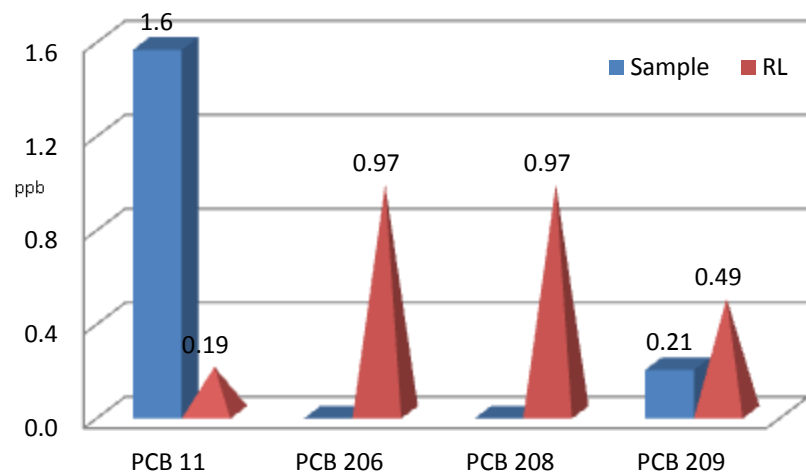
Green lime Jello-container



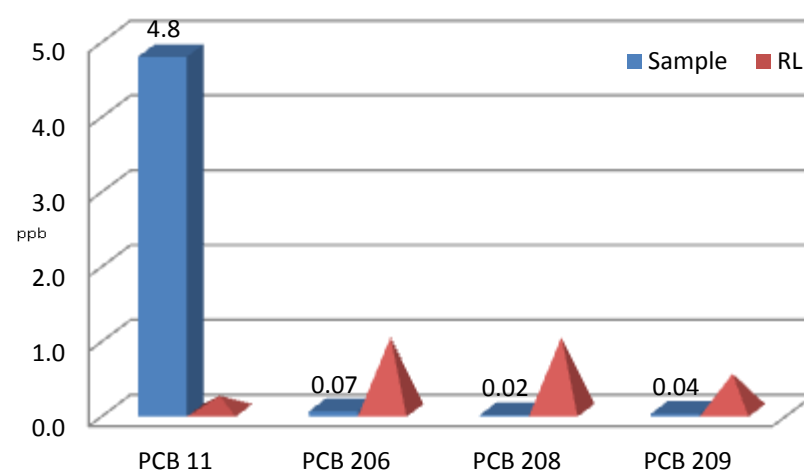
Lemon yellow Jello-container



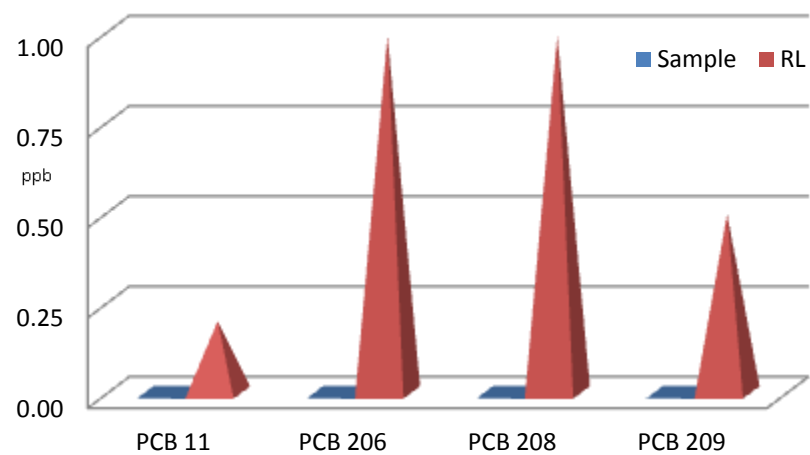
Club crackers-green package



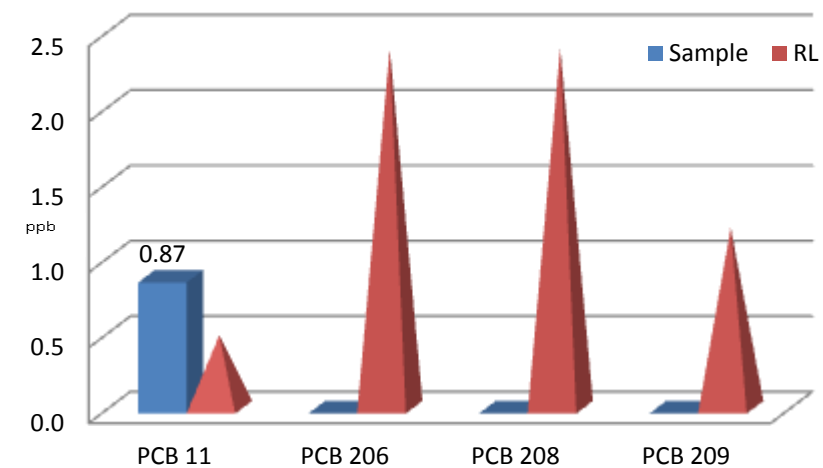
Ancient Grains Cereal-package



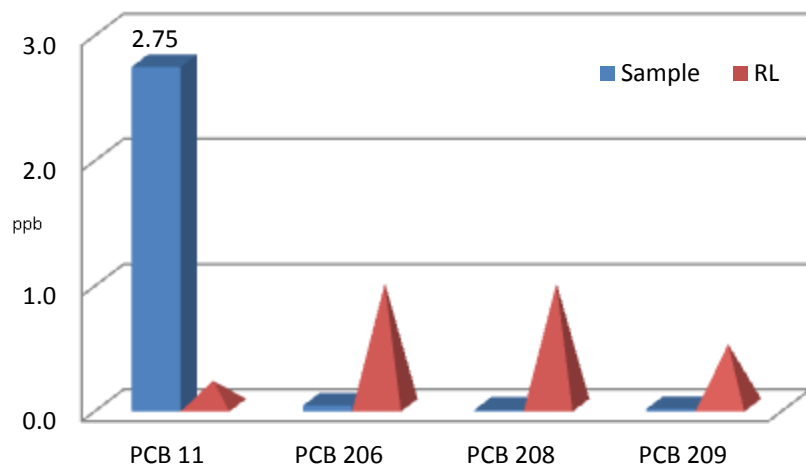
Sun-dried plums-plastic package



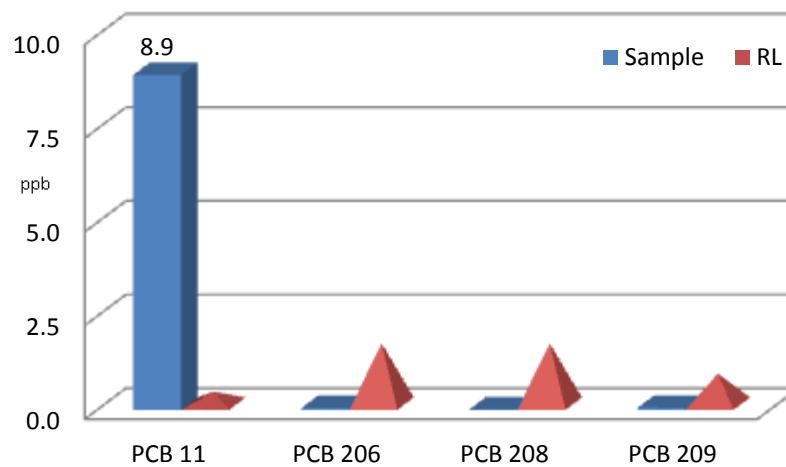
Sun-dried apricots



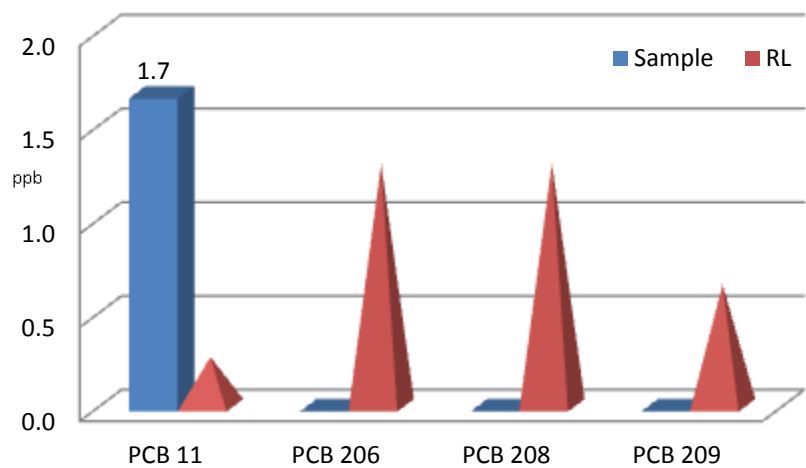
Lemon wafers-package



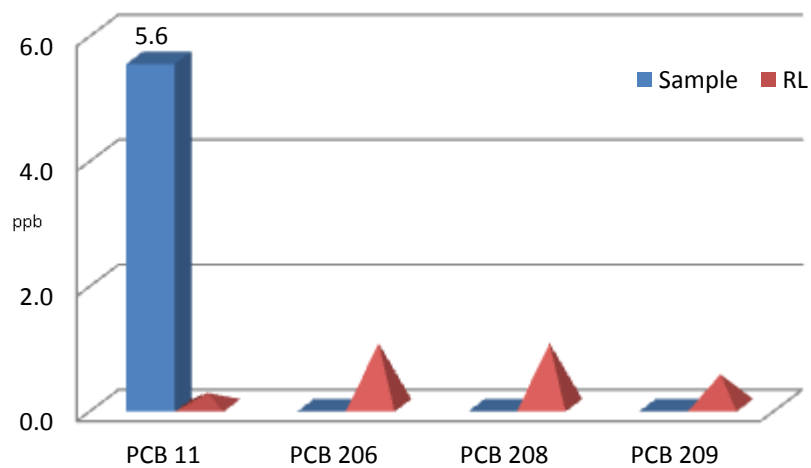
Macaroni and Cheese-package



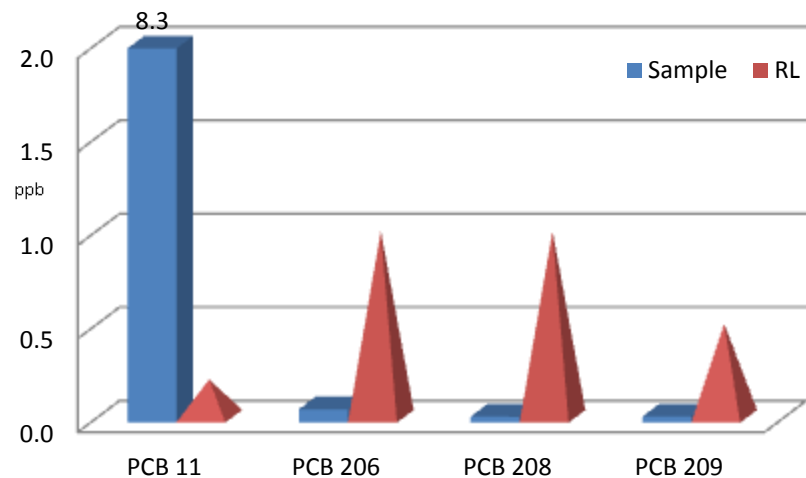
Mild Duds-package



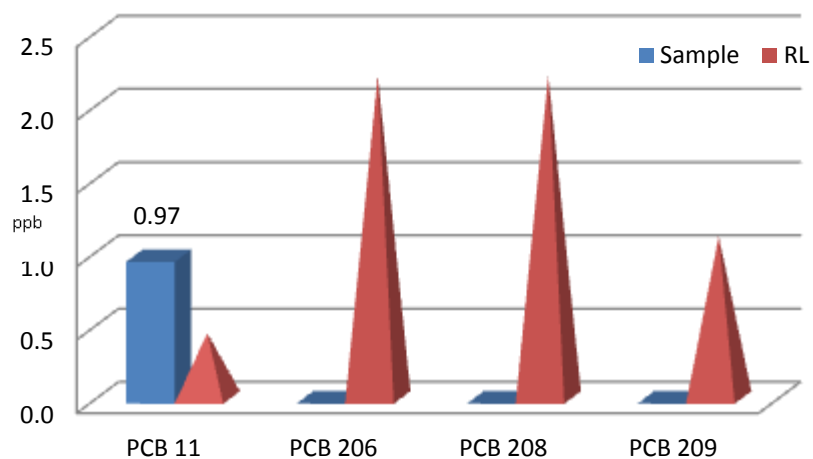
Fruit Thins-plastic container



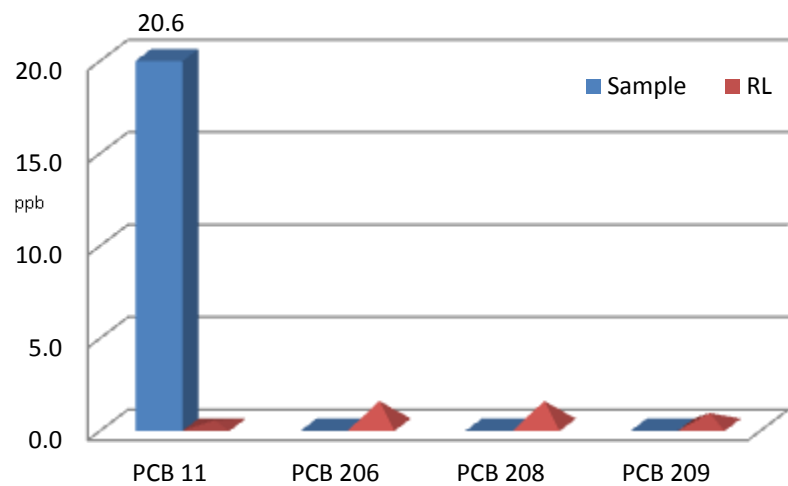
Crunch granola bars-package



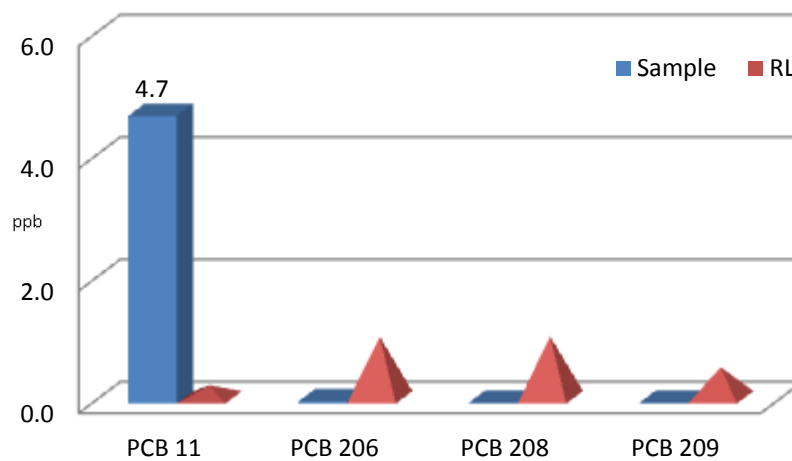
Toll House Chocolate Chip-package



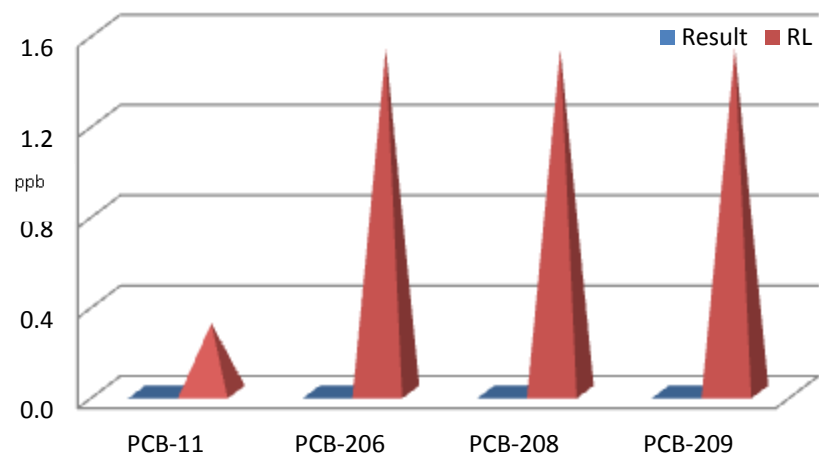
Fruit Thinspackage



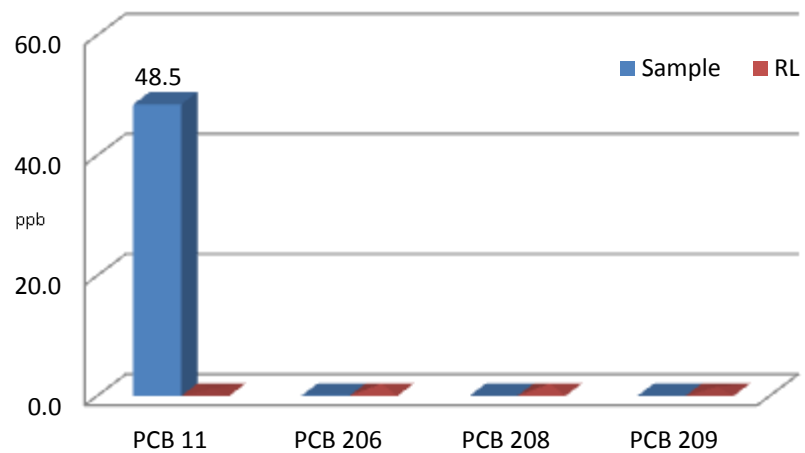
Wafer cookies-package



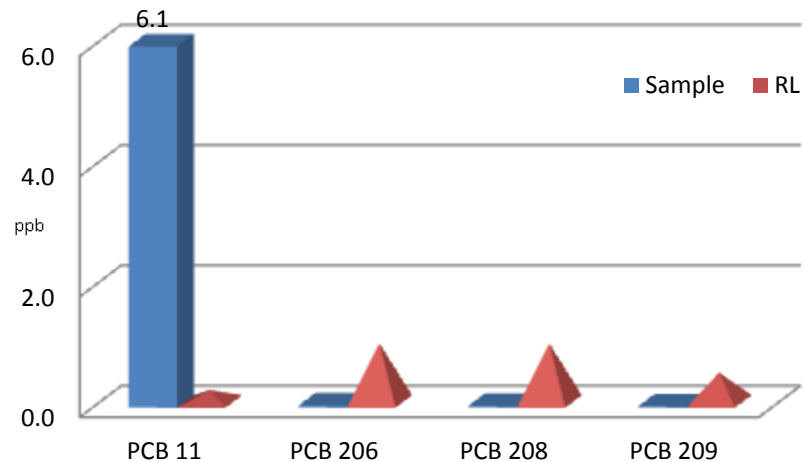
Golden Oreos-packaging



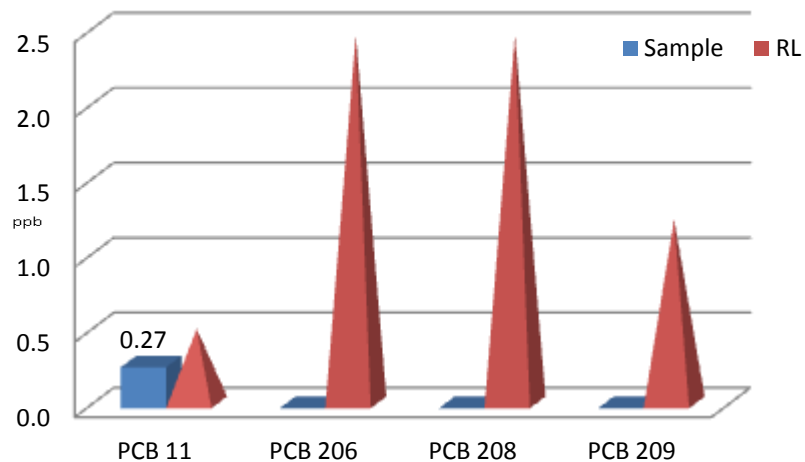
Crackers Handi-Snack-package



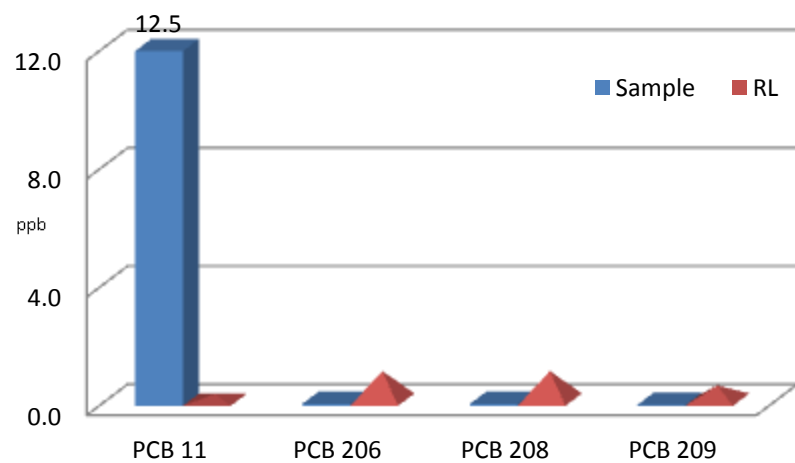
No calorie sweetener-package



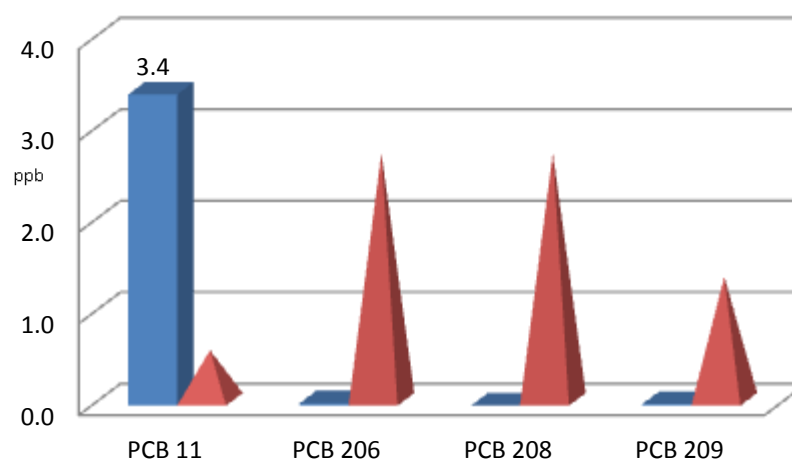
Natural zero calorie sweetener-package



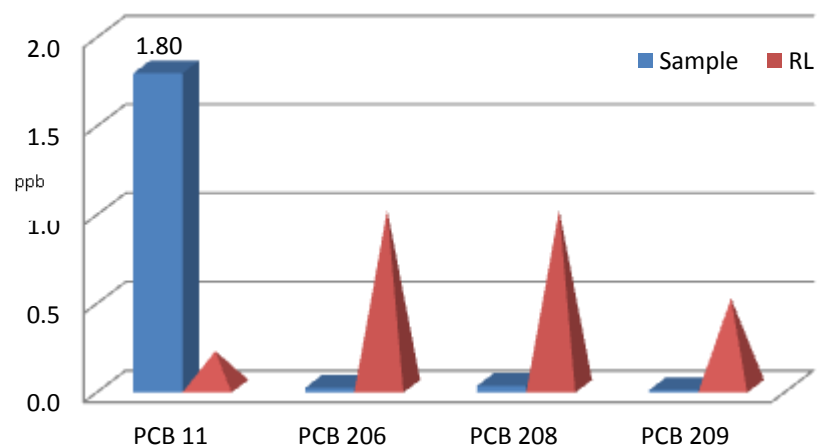
Taco Shells-container



Shells & cheese dinner-package

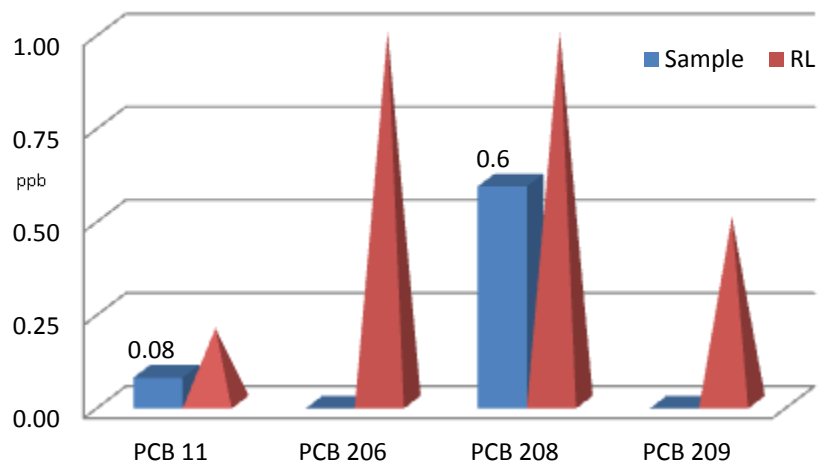


Crackers-container

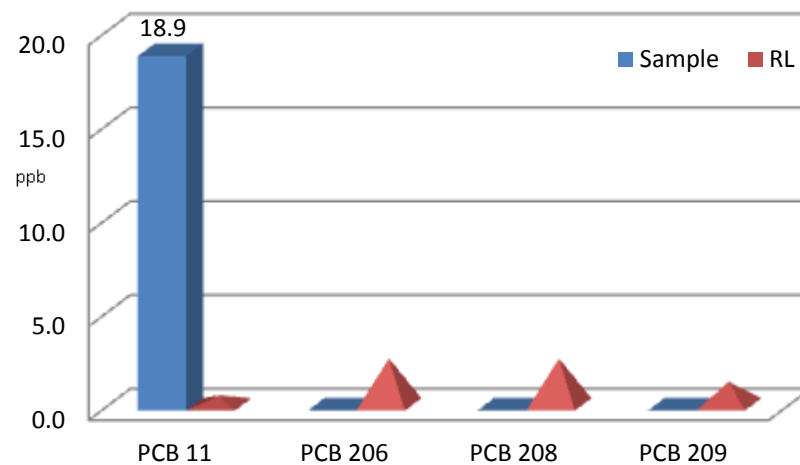


Graphs of individual products - Paper Products

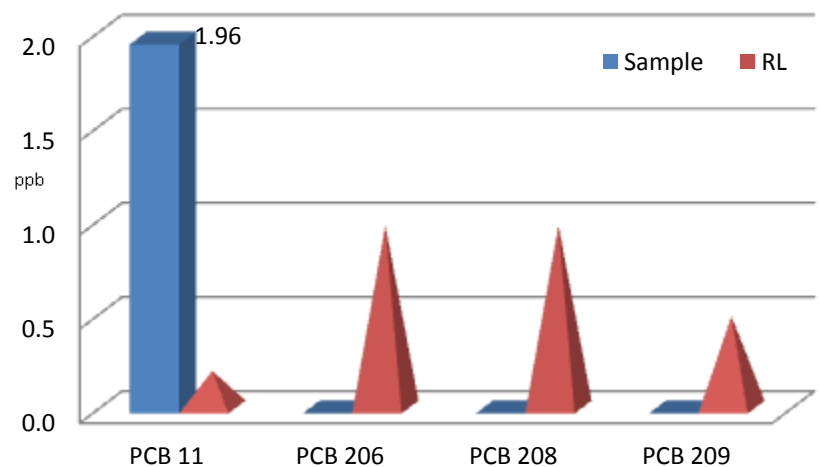
Post-It 3" by 3"



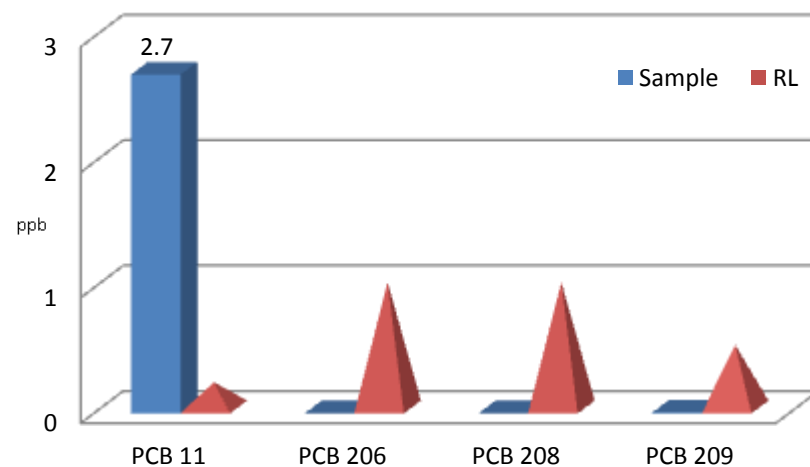
Magazine #1-cover & 1st page



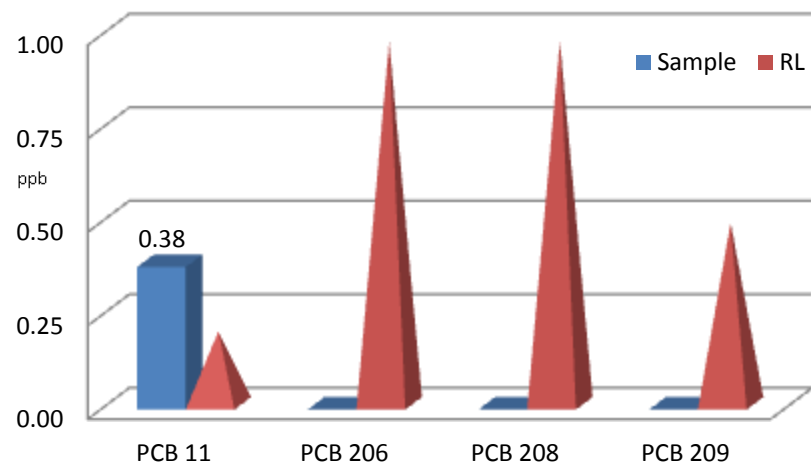
Magazine #2-cover & first 3 pages



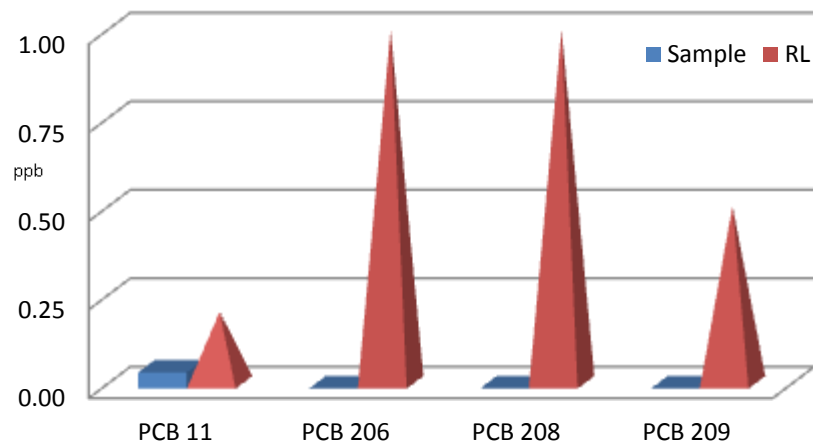
Magazine #3-3 inside pages



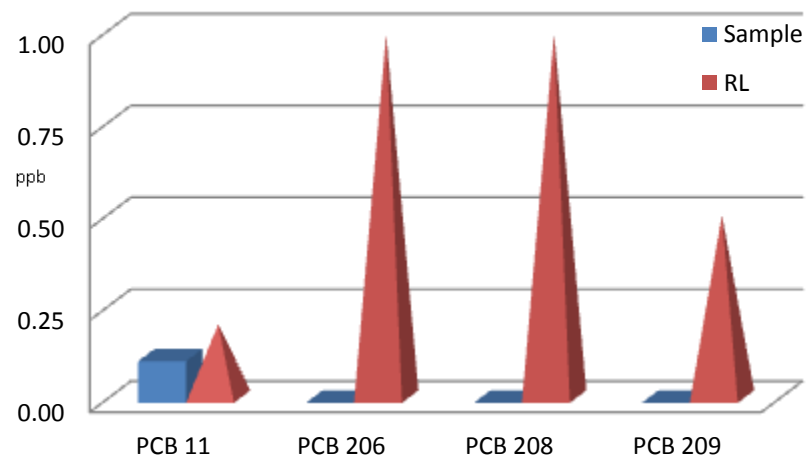
Business meeting handouts



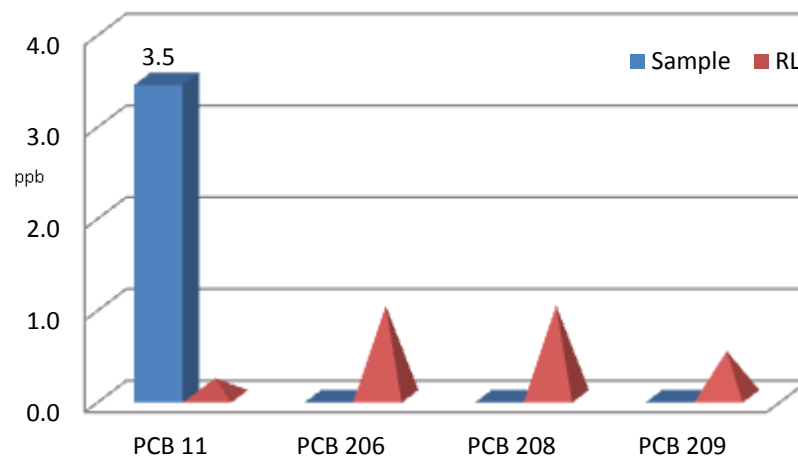
Multi-purpose paper-paper



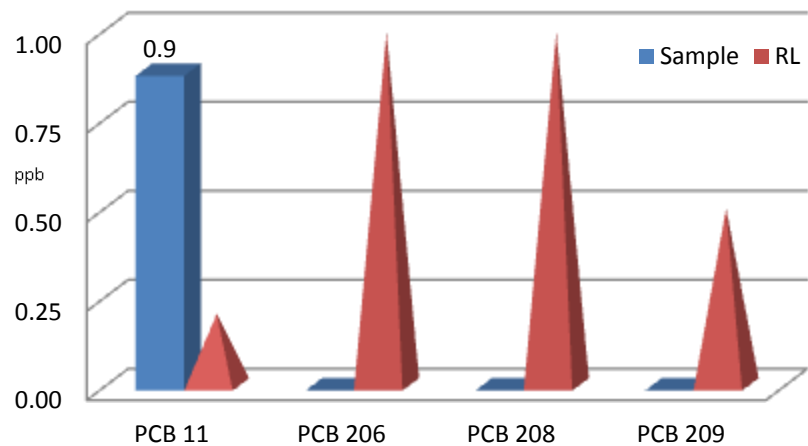
Yellow lined note pad



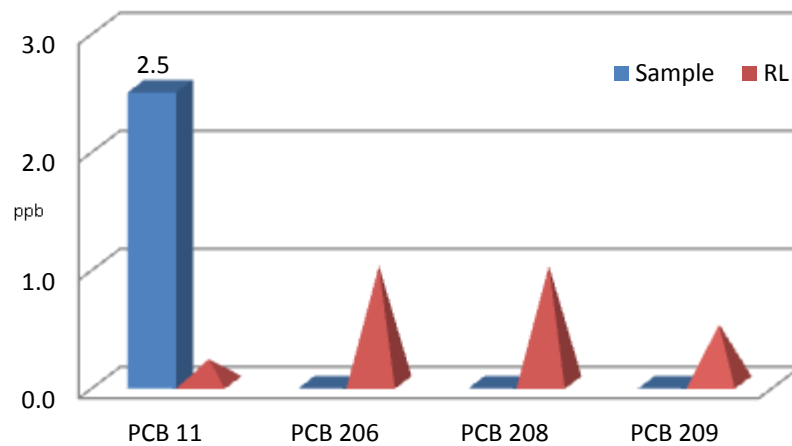
Local newspaper-comics



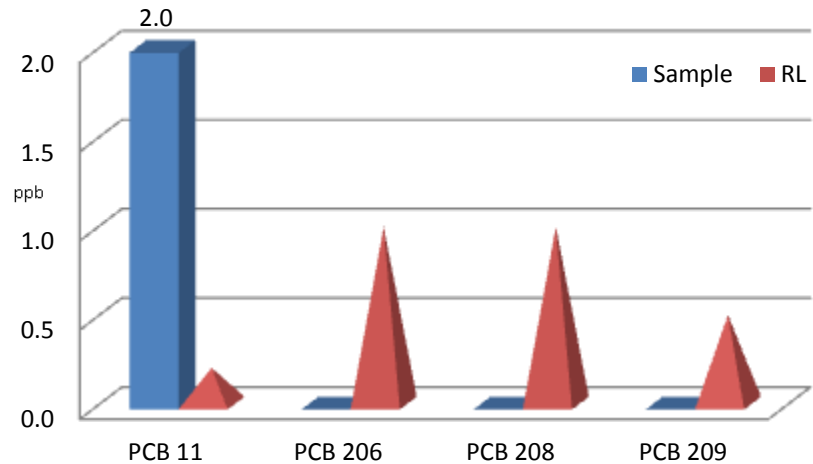
Local newspaper-front page



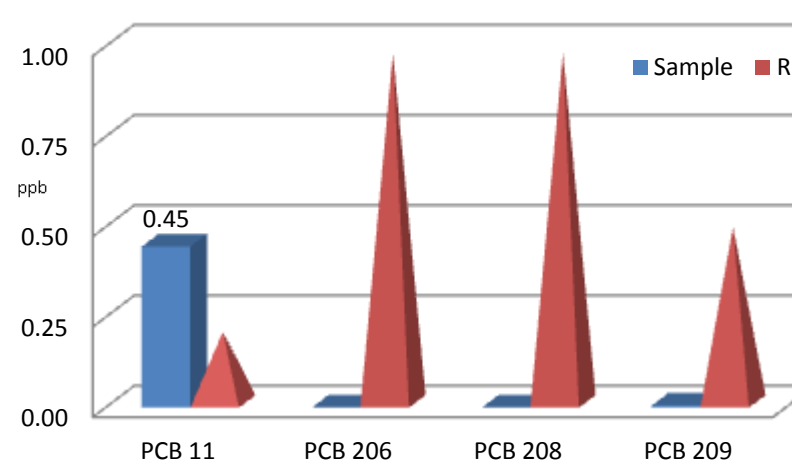
Local newspaper-glossy inserts



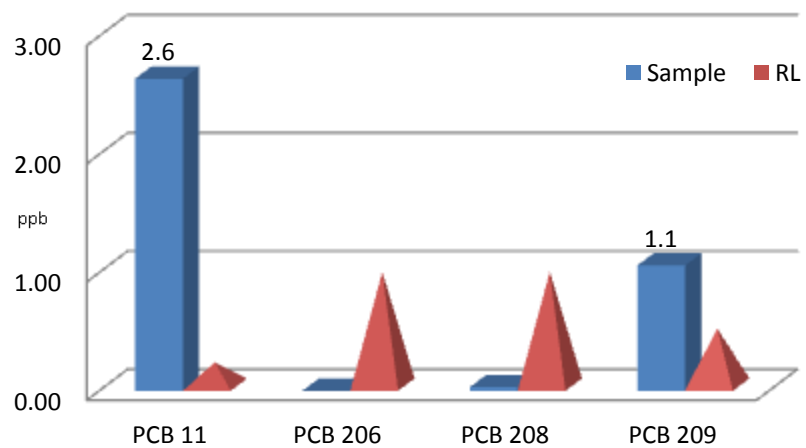
Local paper-non glossy insert



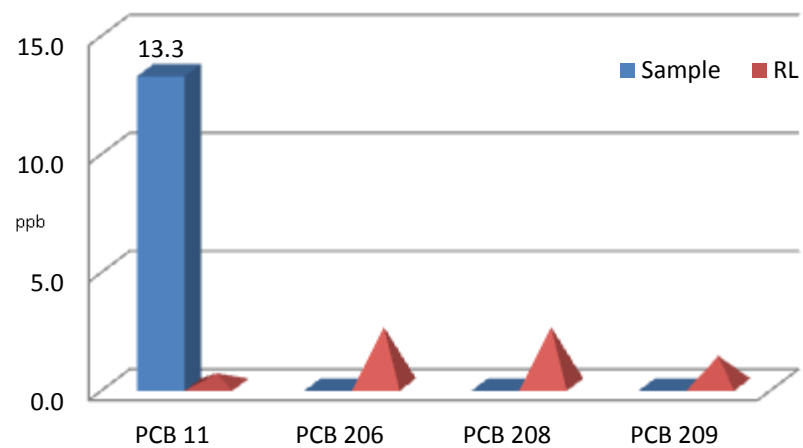
Mag #4-back page w/yellow ad



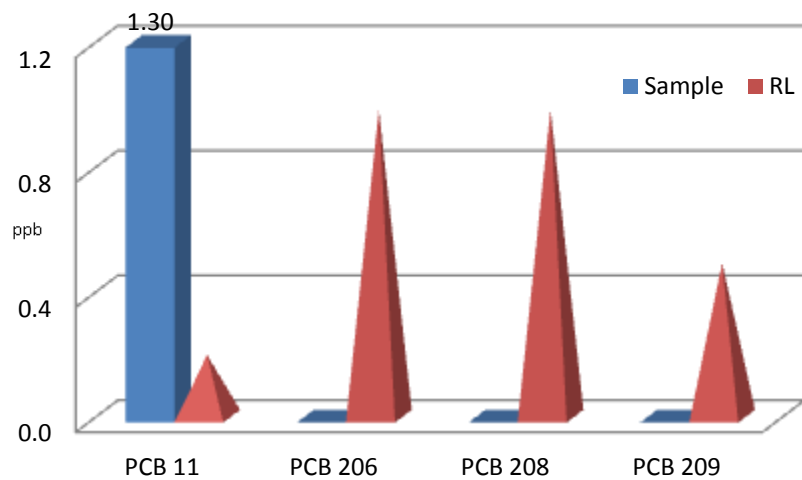
Yellow pocket folders



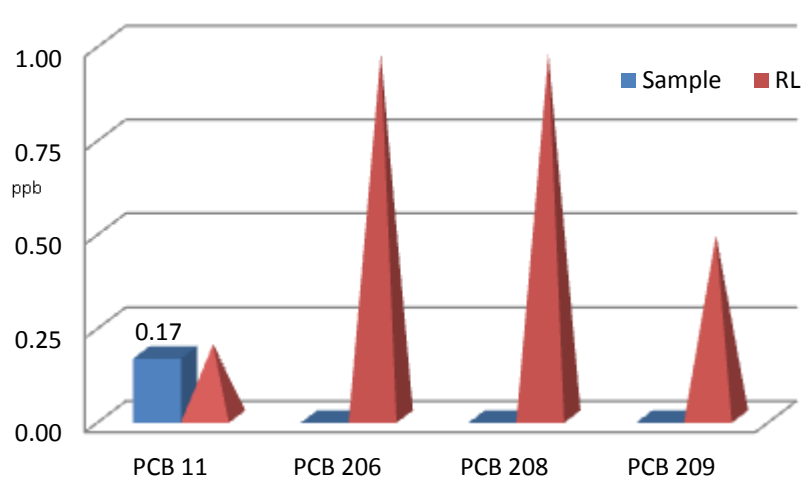
Yellow report dividers

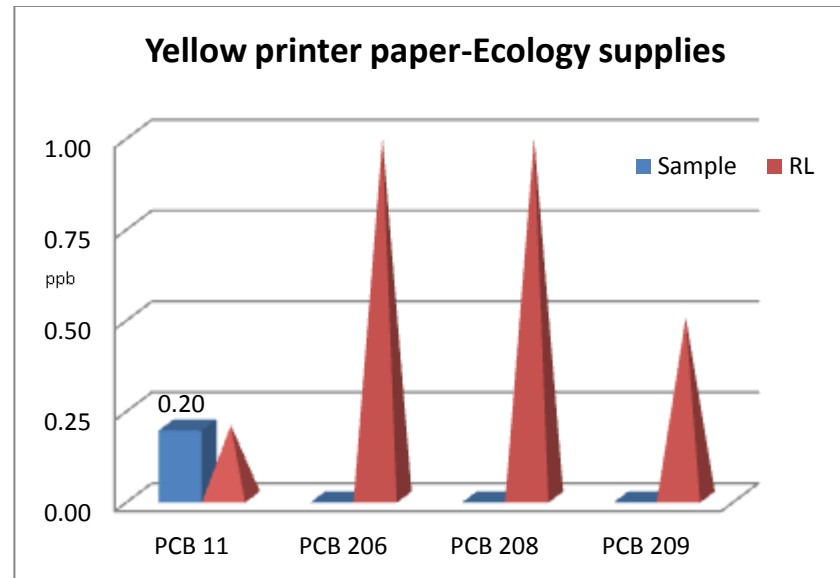


Community College Continuing Ed mail



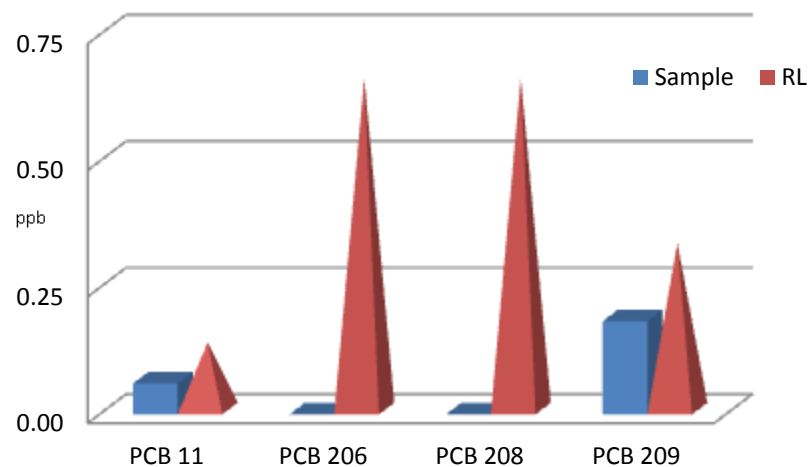
Legal ruled yellow writing tablet-sheets



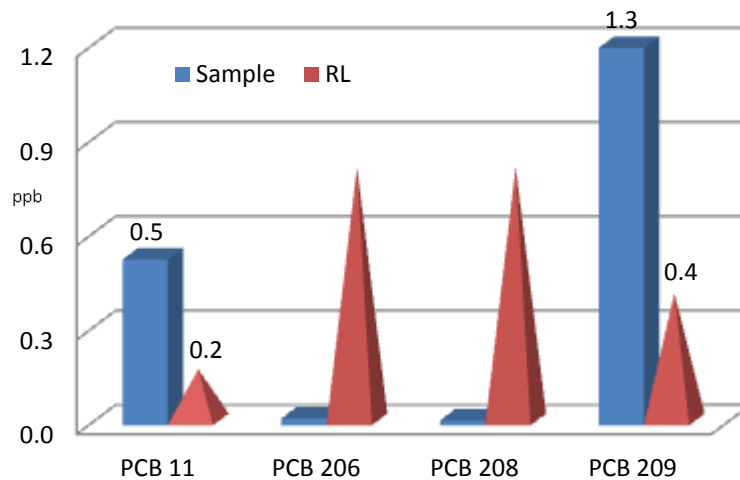


Graphs of individual products - Paints and Paint Colorants

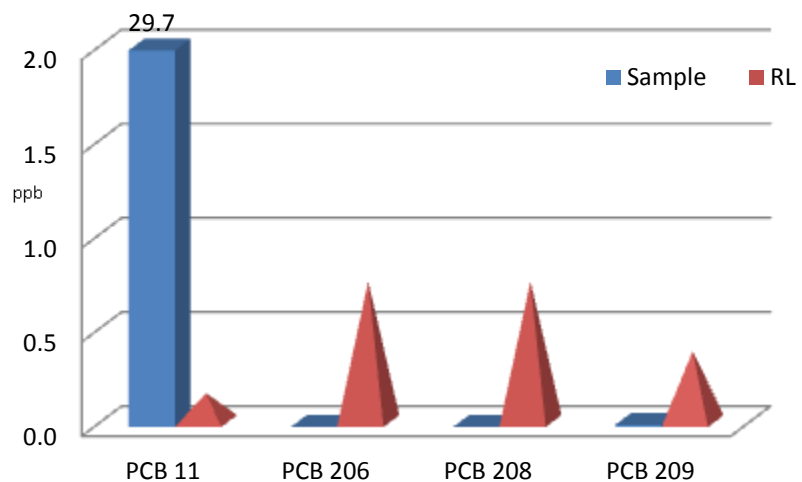
Dirt Fighter paint and primer



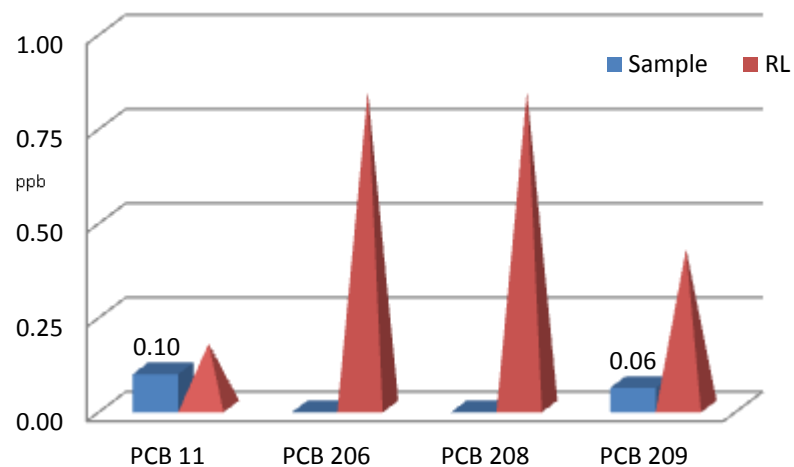
Interior/Exterior Spray Paint-green



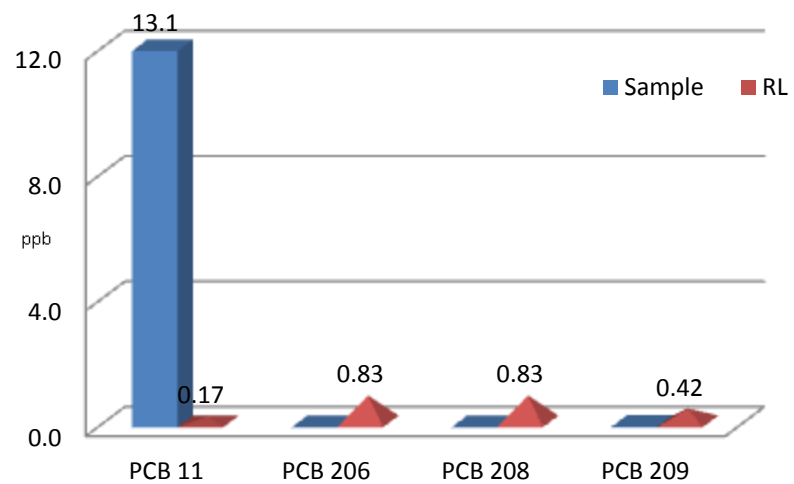
Interior/Exterior Spray Paint-yellow



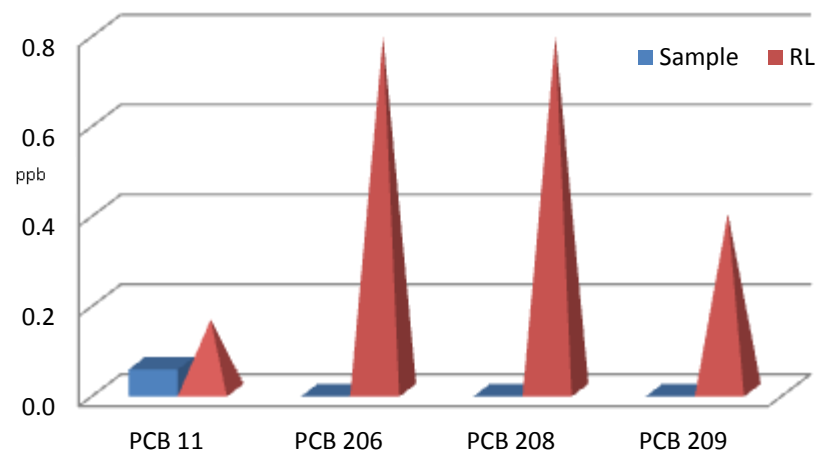
Indoor/Outdoor Blue Gloss spray paint



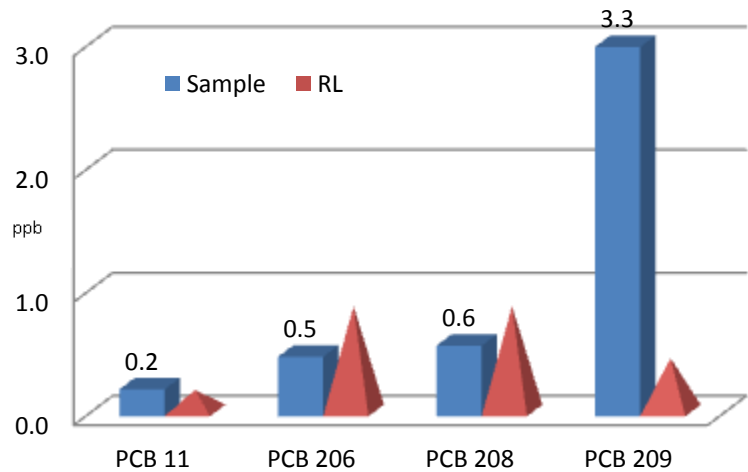
Indoor/Outdoor Yellow Gloss spray paint



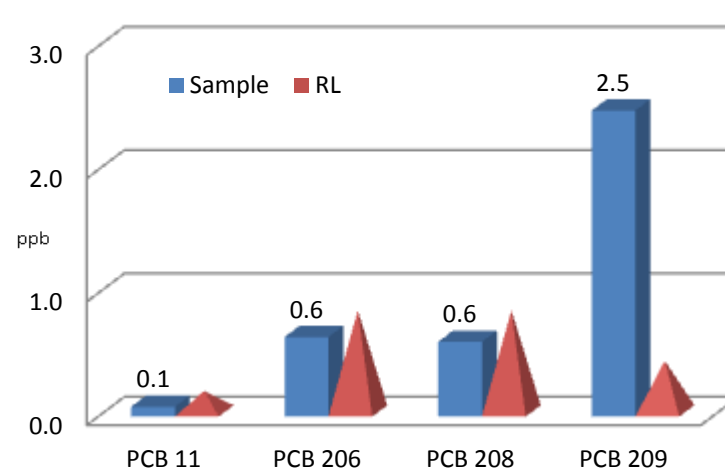
Interior acrylic paint



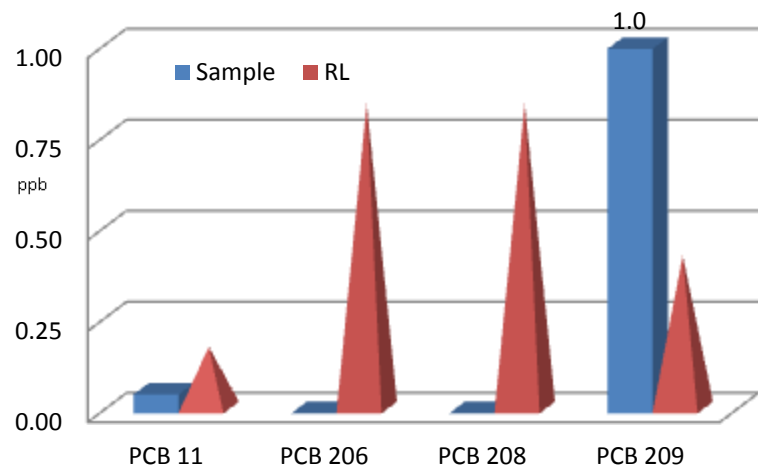
Fluorescent neon green spray paint



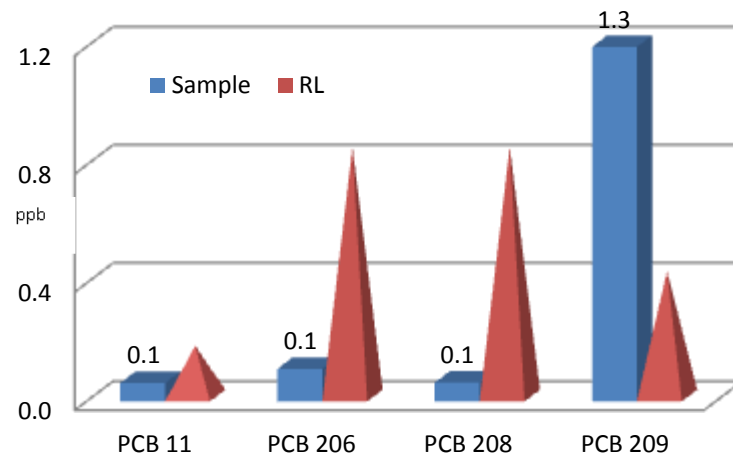
Fluorescent neon yellow spray paint



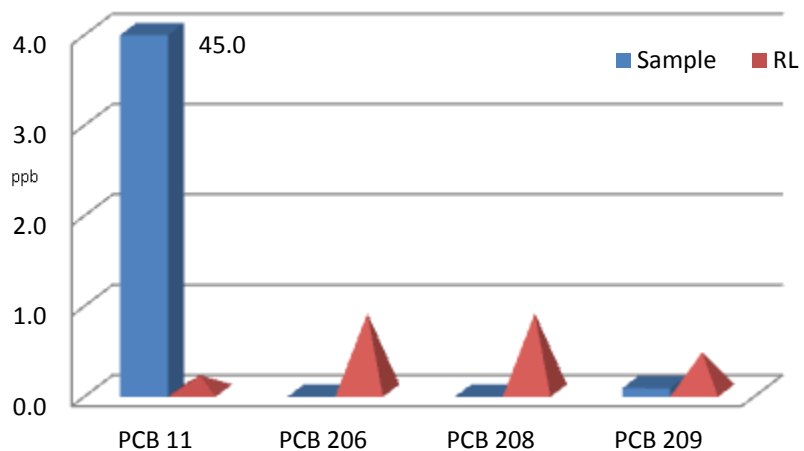
Thalo Green colorants No. 45514Z



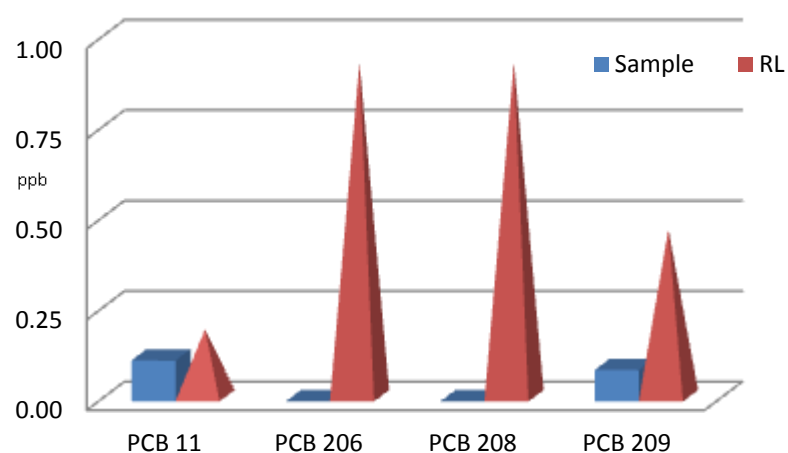
TiO2 low VOC No. 46512Z



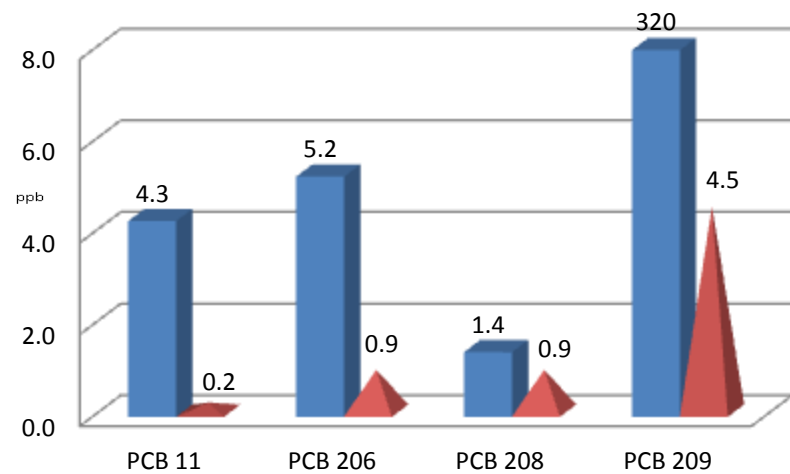
Medium yellow colorants No. 46814Z



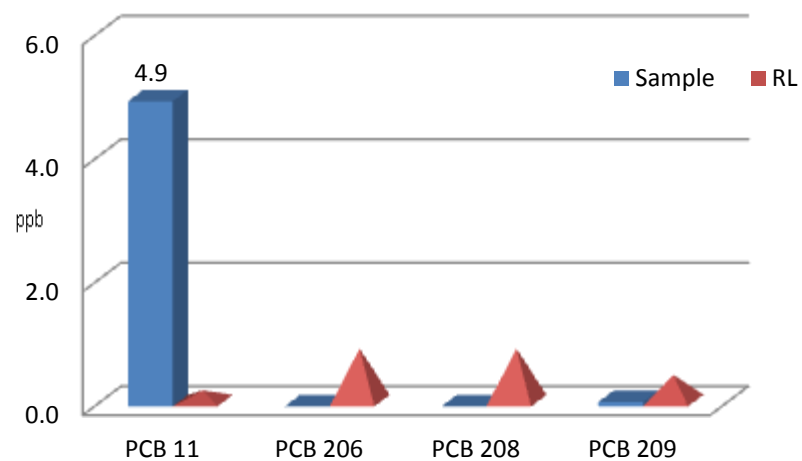
Universal paint colorant-Phthalo blue



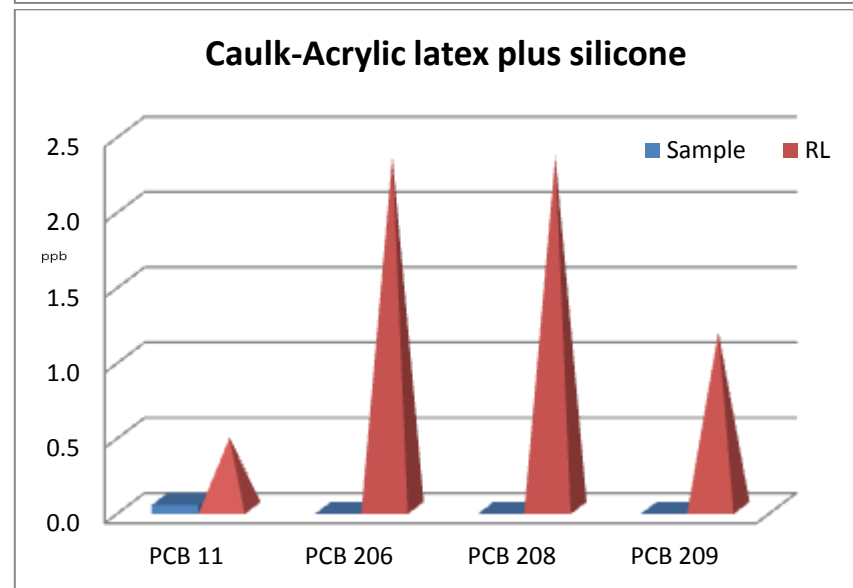
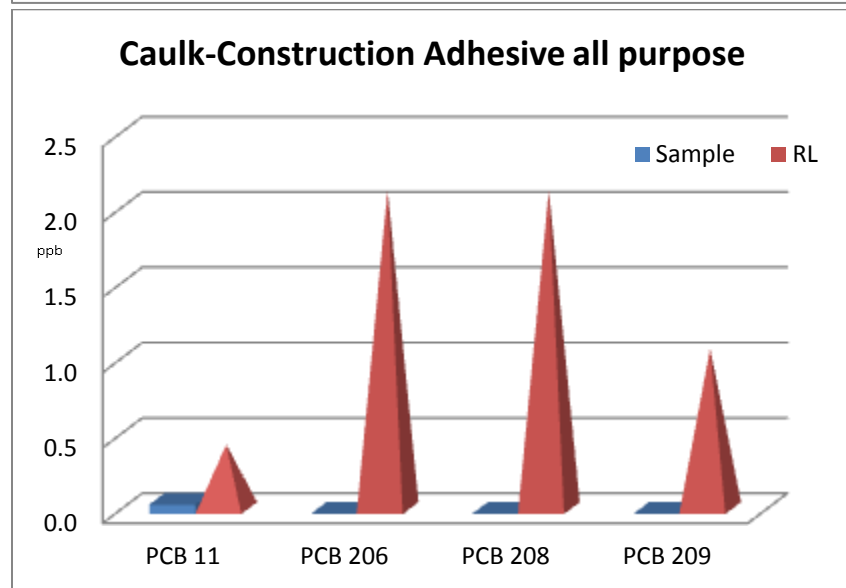
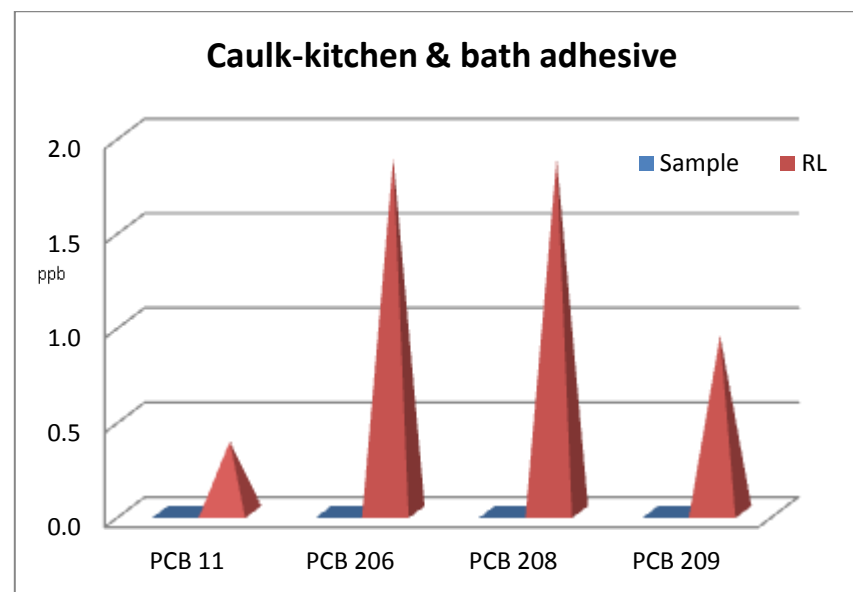
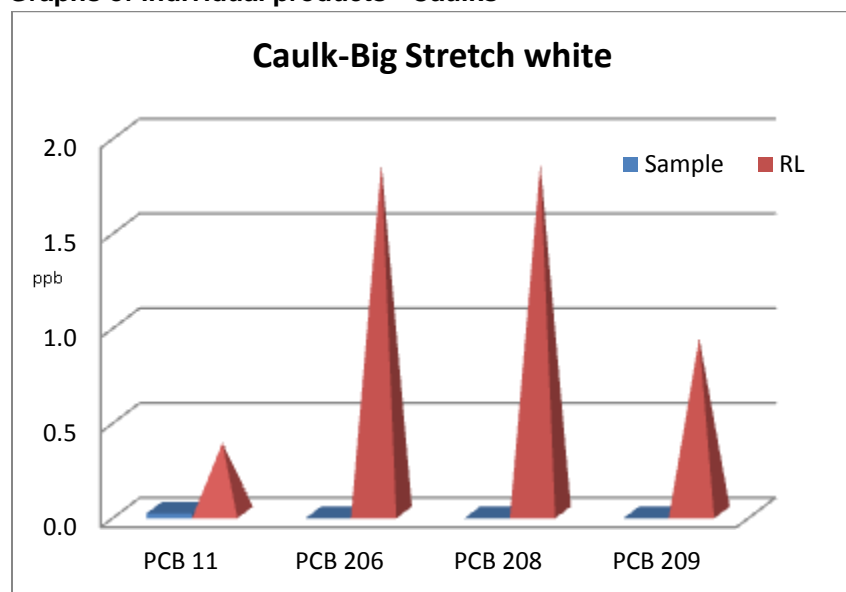
Universal paint colorant-Phthalo green



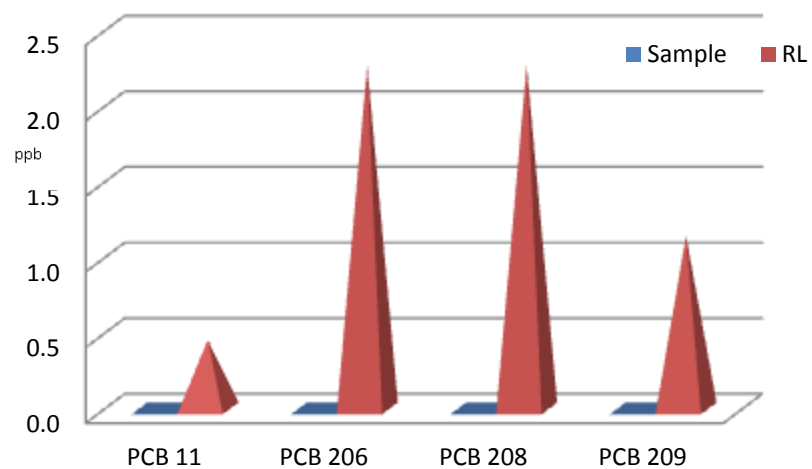
Universal paint colorant-Med. Yellow



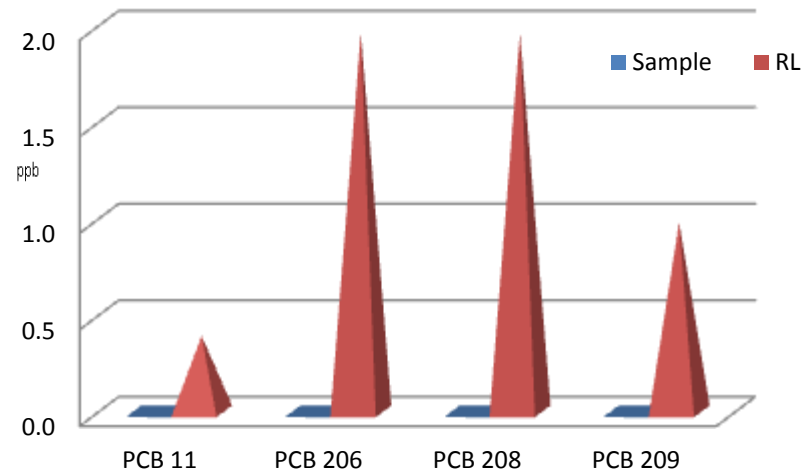
Graphs of individual products - Caulks



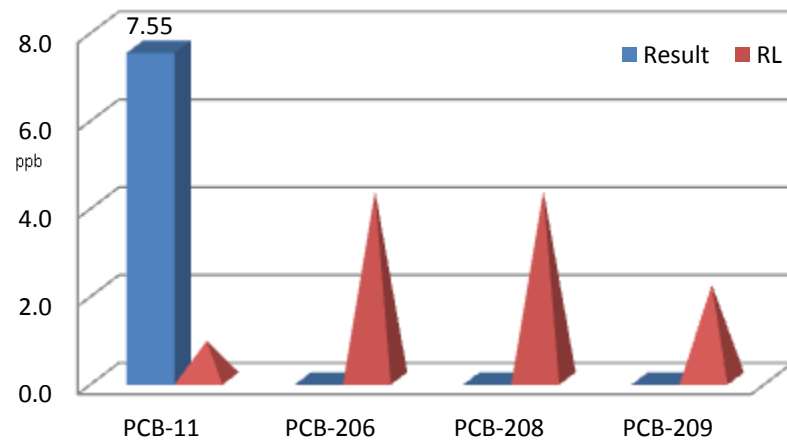
Caulk-Loctite polyseamseal all purpose



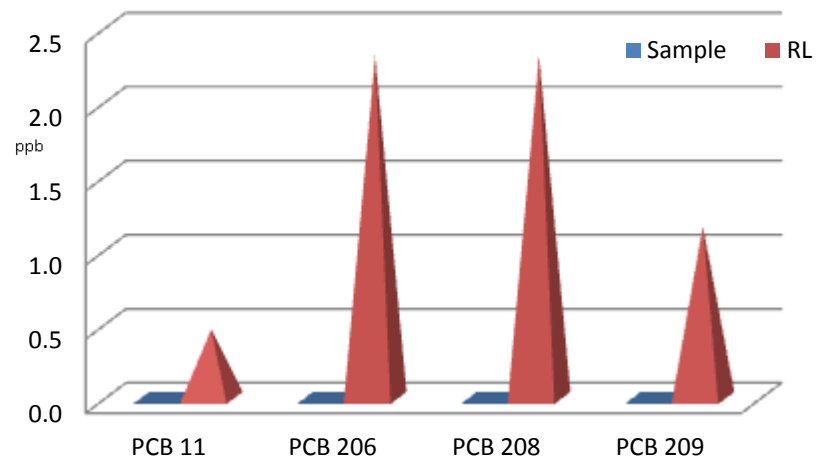
Caulk-Pink 2 White acrylic sealant plus silicone



Caulk-White acrylic plus silicone



Caulk-White Blanco Phenoseal vinyl adhesive



Graphs of individual products - Inks and Food Products

