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James D. Okun

O'Reilly, Talbot & Okun Associates, Inc., okun@oto-env.com

Andy Rezendes

Alpha Laboratory, Inc

James Occhialini

Alpha Laboratory, Inc

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PART II: Indoor Air

Chapter 5

HOW OVERLY CAUTIOUS RISK ASSESSMENT METHODS OVERSTATE RISK FROM PCBs IN INDOOR AIR

James D. Okun^{1§}, Andy Rezendes² and James Occhialini³

¹*O'Reilly, Talbot & Okun Associates, Inc., 19 West Main Street Suite 205, Westborough, MA 01581*, ²*Alpha Laboratory, Inc, 320 Forbes Boulevard Mansfield, MA 02048*, ³*Alpha Laboratory, Inc. 8 Walkup Drive, Westborough, MA, 01581*

ABSTRACT

The past use of building materials that contained polychlorinated biphenyls (PCBs) is prompting public concern and expensive PCB removal projects. Building materials that may contain PCBs include paint, caulk, floor finishes and many other interior and exterior construction components manufactured before 1972. While initial concern about PCBs in buildings has focused on schools, it is likely that PCB-containing materials will also be found in many residential, commercial, and industrial structures.

In buildings, the primary route of human exposure is the inhalation of PCBs that volatilize out of building materials. The USEPA has indicated that inhalation of airborne PCBs may pose a significant human exposure pathway in schools. EPA's approach to PCBs in schools is evolving quickly. Initial estimates of health risk from indoor PCBs were calculated using risk assessment methods and EPA published toxicity factors. Recently, EPA established guidance titled "Public Health Levels for PCBs in Indoor School Air" to assist school systems in remediation efforts. These criteria are intended to be "prudent public health levels that maintain PCB exposures below the 'reference dose' – the amount of PCB exposure that EPA does not believe will cause harm."

[§]Corresponding Author: James D. Okun, O'Reilly, Talbot & Okun Associates, Inc., 69 Milk Street Suite 218, Westborough, MA 01581, (508)366-6409, okun@oto-env.com.

This article considers two factors central to the accurate assessment of PCB indoor air risk: 1) the difference in the chemical make up of PCBs in air compared to the chemical make up of the PCBs in the building materials they originated from; and 2) how the variability in certain commercial lots of PCBs (Aroclors) can result in different degrees of toxicity. Each of these factors may act to significantly modify the level of risk associated with the inhalation of PCBs in buildings. At the present time, EPA has not incorporated these factors into its calculation of its Public Health Levels.

Keywords: PCBs, PCBs in building materials, PCBs in schools, PCBs in indoor air, Aroclor 1254.

1. INTRODUCTION

The work described in this article began as research to better understand how the chemical makeup of PCB mixtures change as they volatilize out of a solid matrix and into air. This topic is of interest because the inhalation of PCBs in indoor air is increasingly seen as a significant exposure pathway for building users. Commercially manufactured PCBs were mixtures of many different but similar chemicals with varying degrees of chlorination. The volatility of different PCBs is generally correlated with their degree of chlorination, such that PCBs with more chlorine substitutions are less volatile than those with fewer chlorine substitutions (Fiedler, 2002; Foreman and Bidleman, 1985). Available research supports the notion that PCBs with less chlorination preferentially volatilize out of solid and/or liquid media (Casey et al, 1999).

There is also a general correlation between the degree of a PCB mixture's chlorination and its level of toxicity (USEPA, 1996). It has been observed that PCB mixtures with lower average molecular weights (and thus a lower degree of chlorination) are less toxic than those with higher average molecular weights (and thus a higher degree of chlorination). Therefore, the partitioning process that occurs when PCBs volatilize into air may result in the airborne PCBs having a lower degree of chlorination and a lower degree of toxicity than the parent PCB mixture in the solid source materials.

Based on the review of almost 300 indoor air sample PCB test results considered in the present study, Aroclor 1254 was the most frequently detected commercial PCB product among the samples. Further research into the chemical makeup of Aroclor 1254 revealed that, while there is batch to batch variability, there were also two broadly different 1254 products with different chemical and likely toxicological characteristics. These two Aroclor 1254 types appear to have

arisen from different manufacturing processes and are likely have different toxicological properties.

1.1 PCBs in Building Materials and Their Initial Regulation

There has been growing attention to the past use of PCBs in building materials because of their ability to volatilize from these materials into indoor air (Herrick et al. 2004; MIT, 2007; and MADPH, 2009). Prior to 1971, PCBs were used as ingredients in paints, caulk and adhesives to impart plasticity and extend the useful life of the materials to which they were added. Sometime in the 1970-1971 period, Monsanto Company, the sole US PCB manufacturer, stopped the sale of PCBs for uses that were not considered “totally enclosed”. Since the use of PCBs in building materials was not considered totally enclosed, it is likely that the manufacture of PCB containing building materials stopped after this time. The concept of limiting future uses of PCBs to those that were “totally enclosed” was carried through to the 1976 Toxic Substances Control Act (TSCA, 1976) and from there to the 1978 PCB regulations promulgated pursuant to TSCA.

The 1978 PCB regulations (40 CFR 761) brought a legal end to use of PCBs in new building products, but did not place limitations on the continued use of existing PCB containing building materials. EPA’s initial PCB enforcement activity focused on: 1) stopping the production and continued distribution of new PCBs in commerce; and 2) taking steps to identify and regulate the remaining stock of liquid PCBs and PCB electrical equipment. The bulk of PCB liquids were in electrical equipment such as transformers and capacitors, and EPA believed the greatest PCB risks arose from the potential mismanagement of these PCB liquids.

With the promulgation of 1978 PCB regulations, the continued legal use of existing PCBs could only occur when permitted by specific “use authorizations” described in the regulations. While different from “use authorizations”, the 1978 regulations also contained a *de facto* exemption for PCBs that were defined as being “in service”. The “in service” category was a regulatory catch-all that lumped together a variety of PCBs whose continued use was considered outside of the intended scope of the PCB regulations.

The “in service” category included the continued use of existing, in-place PCB-containing building materials. However, the “in service” category also included PCBs that had been released to the environment prior to the effective date of the regulations; this was because the TSCA regulations were not intended to be used to enforce the cleanup of pre-1978 PCB releases. In 1978 this was not seen as a regulatory deficiency because the agency had other legal tools to force cleanups of spilled PCBs.

When the 1978 PCB regulations were adopted, there were few health or environmental concerns being expressed about PCBs in building materials. This reflected the view that the PCBs in building materials were effectively “locked in” to the materials and, thus, did not pose an exposure risk.

1.2 The 1998 Mega-Rule and Increased Regulation of PCB Containing Building Materials

In 1998, EPA promulgated sweeping revisions to the PCB regulations; so many changes were made that the new regulations were referred to as the “1998 PCB Mega-Rule”. The preamble to the Mega-Rule explains that EPA had decided to eliminate the concept of PCBs being “in-service” because the agency wanted the ability to regulate pre-1978 PCB cleanups under the PCB regulations. The Mega-Rule contained no explanation or analysis of the fact that by eliminating the “in-service” category the agency was, with the same stroke, making the continued use of PCBs in existing building materials a TSCA violation.

Following the 1998 PCB Mega-Rule changes, EPA seemed slow to recognize that it had adopted the responsibility for regulating in-place PCB-containing building materials. This slow recognition could be seen in the absence of EPA's program planning and inconsistent regional responses to PCBs in building materials. Recently, national guidance on the management of PCB-containing building materials has begun to appear on EPA's web site.

1.3 PCBs in Indoor Air from Building Materials

By the early 1990s, reports of PCBs in indoor air were appearing in research journals. Much of the early work was conducted in Germany (Benthe et al, 1992; Balfanz et al, 1993). A survey article on indoor air PCB testing appears in *Spengler's Indoor Air Quality Handbook* (Spengler et al, 2000). In the subsequent eleven years, there have been a growing number of articles describing PCBs in building materials and their potential to enter indoor air, particularly in schools (Daley, 2009; Egbert, 2008). The greater frequency of PCB detections in schools is in part the result of schools being the most common location for investigators to examine.

Initial estimates of health risks from PCBs in indoor air were based on the use of standard EPA exposure and risk assessment models. These assessments considered carcinogenic and non-carcinogenic risk. More recently, EPA has published “Public Health Levels for PCBs in Indoor School Air” that are based on the Aroclor 1254 PCB reference dose (RfD) that uses solely a non-cancer toxicological endpoint (USEPA, 2009).

2. MATERIALS AND PROCEDURES

The term PCB does not refer to single chemical, but rather to a group of chemicals with between one and ten chlorine substitutions that all have the biphenyl 12-carbon aromatic chemical structure in common. There are 209 different chemical arrangements of chlorine atoms that can occur on biphenyl; each of these different chlorinated biphenyl arrangements is referred to as a “congener”, and there are 209 different congeners in the PCB group. Different congeners that have the same number of chlorine substitutions are referred to as “homologs”; there are ten different homolog subgroups ranging from the monochloro homolog (with one chlorine substitution) to the decachloro homolog (with ten chlorine substitutions).

The goal of commercial PCB manufacturing was the production of congener mixtures rather than making any single congener. The sole US producer of PCBs was the Monsanto Chemical Company and its PCB products were sold under the trade name “Aroclor”. Each Aroclor PCB mixture contained many different congeners. There were several different formulations of Aroclors that were differentiated based on the percent chlorine in the mixture (e.g. Aroclor 1242 would have 42% chlorine and Aroclor 1254 possessed 54% chlorine). Of the 209 different PCB congeners, a number of them occur rarely or not at all in Aroclors mixtures; these rare or absent congeners are referred to as non-Aroclor PCBs.

2.1 Measurement of PCBs in Media

Most analyses of PCBs employ: 1) an extraction step where PCBs are transferred from the sample to an organic solvent; 2) a separation step where the sample extract is introduced on to a gas chromatograph; and 3) a detection step where the presence of the PCBs is identified and recorded, a variety of detectors may be used for this step. Depending upon the specific project requirements, the results of PCB analysis may be quantified as Aroclors, as homologs, or as individual congeners (in increasing order of analytical difficulty).

When there is little likelihood of change in the makeup of a PCB mixture in a sample matrix due to environmental weathering or other influences, analysis of samples for Aroclors often proves adequate. However, if there is reason to suspect that the PCB composition has changed, or if the goal of the project is to assess possible changes in PCB makeup, then analysis for PCB homologs or congeners will produce more useful results (Prignamo et al, 2006). Quantification of PCBs as Aroclors presumes that the different PCB components (congeners and homologs) are present in fixed ratios to each other, whereas analysis for homologs and congeners does not require this assumption.

Since the objective of the present investigation was to assess whether the composition of PCBs changed as they volatilized out of the solid phase (building materials) into the gaseous phase (indoor air), it was necessary to compare the results of congener and homolog data for the solid phase to that of the air phase.

2.2 Measurement of PCBs in Air

To conduct a meaningful comparison of indoor air PCB composition to the PCB composition in building materials, it was necessary to acquire a PCB in air data set that represented a variety of different settings. Two of this article's authors (Rezendes and Occhialini) are engaged in the day to day analysis of air samples for PCBs and they provided the results of approximately 300 indoor air samples for PCBs quantified as either congeners or homologs. Air samples were collected in accordance with EPA Method TO-4/TO-10 and were analyzed in accordance with EPA Method 680/8270-SIM (USEPA – SW846, 2011).

There were two limitations inherent to this indoor air data set: 1) because of the need to maintain confidentiality regarding the sampled location, the only information that could be made available for this work was the mass of each congener or homolog measured on the sample cartridge; and, 2) it was not possible to directly compare the indoor air PCB result with a known solid phase PCB source.

2.2.1 Sample Result Limitations

Because the mass of each congener or homolog retained on the sample cartridge was the only information available for each sample, it was not known what the actual concentration of each PCB was in air. In other words, the sample volume was not known. This was not a serious shortcoming because the current study was concerned with the relative abundance of each congener or homolog as a fraction of the total mass of PCBs in the sample. Not knowing the volume of air collected for the sample did not limit the usability of the data for assessing the composition of PCBs in the sample.

2.2.2 Limitations Due to Inability to Compare Air Data with a Known PCB Source Material

The inability to compare air data with a known PCB solid phase source was potentially a more serious limitation. The overall study objective was to assess whether there were changes in the mixture composition as the PCBs volatilized out of the solid phase source and into indoor air. Not knowing the composition of the starting material was an obstacle. However, in most cases it was possible to identify the parent Aroclor (that is the Aroclor present in the solid phase) based on

the congeners/homologs detected in the air samples. Knowledge of the Aroclor type was generally sufficient to develop an approximation of the starting material PCB composition.

In fact, there is documentation in the literature supporting the general uniformity of homolog distribution in the Aroclors. There is more batch to batch variability in the congener makeup of the Aroclors than there is homolog variability. Therefore, PCB homolog distribution results were chosen as the comparative parameter that would be used in this study. For the purpose of this work, the Aroclor homolog distributions published by Frame (Frame et al, 1996) were adopted as the basis for comparison to the air samples.

3. DATA, ANALYSIS AND DISCUSSION

To permit a comparison of air sample PCB homolog results to the results for Aroclors, three indicators were used along with a more general evaluation of the homolog pattern for the samples and Aroclors.

3.1 Homolog Data for Aroclors

The PCB Aroclors have distinctive homolog profiles that make identifying them from homolog data straightforward (see Table 1). As a primary identifying characteristic, each Aroclor has a different dominant homolog (except for Aroclors 1242 and 1016) and a second identifying characteristic is the relative concentration of the next heaviest homolog. These two characteristics are generally adequate for identifying an Aroclor from homolog data. A variation of a few percent in Aroclor homologs from batch to batch is not uncommon.

Another indicator that is a useful characteristic of a PCB mixture is the average number of chlorine substitutions per PCB molecule (last column in Table 1). This value is calculated with the following formula:

$$CI/PCB = 1 * (\text{percent mono}) + 2 * (\text{percent di}) + 3 * (\text{percent tri}) + \dots + 10 * (\text{percent deca})$$

The CI/PCB indicator can be helpful for comparing homolog data from environmental samples to parent Aroclors. Changes in the degree of chlorination of a mixture can be tracked using this statistic.

Table 1. Homolog Content and Average Chlorine Substitutions per PCB molecule for Aroclors

	mono	di	tri	tetra	penta	hexa	hepta	octa	nona	deca	Totals	Cl/PCB
1221	60.05%	33.27%	4.20%	1.89%	0.48%	0.00%	0.00%	0.00%	0.00%	0.00%	99.89%	1.49
1232	27.98%	26.84%	21.09%	16.80%	2.90%	0.19%	0.00%	0.00%	0.00%	0.00%	95.80%	2.28
1016	0.73%	17.74%	54.61%	26.50%	0.57%	0.00%	0.00%	0.00%	0.00%	0.00%	100.15%	3.09
1242	0.47%	13.86%	48.02%	32.76%	5.48%	0.18%	0.00%	0.00%	0.00%	0.00%	100.77%	3.32
1248	0.02%	0.32%	22.00%	56.49%	17.55%	1.95%	0.57%	0.00%	0.00%	0.00%	98.91%	3.96
1254	0.02%	0.18%	1.24%	16.38%	52.95%	26.75%	2.65%	0.04%	0.04%	0.00%	100.25%	5.14
1260	0.01%	0.00%	0.00%	0.38%	8.07%	41.15%	39.72%	7.94%	0.40%	0.00%	97.67%	6.34
1262	0.04%	0.16%	0.47%	0.43%	3.08%	26.60%	47.34%	19.51%	1.77%	0.00%	99.40%	6.82

* Adapted from Frame et al, 1996; **Bolded** values indicate dominant homologs.

3.2 Homolog Data for Air Samples

The volume of data involved precludes the presentation of the PCB in air data set used for this article. However, some summary statistics are described. Detected PCBs ranged from 2.7 to 6,836 nanograms per sample. Figure 1 shows the distribution of total PCB mass per indoor air sample. In general, where the total mass of PCBs detected in a sample was less than 10 nanograms, reliable homolog information could not be discerned. The most frequently detected homolog (at highest percent concentration) was the penta-chloro homolog, with the tetra-chloro homolog second.

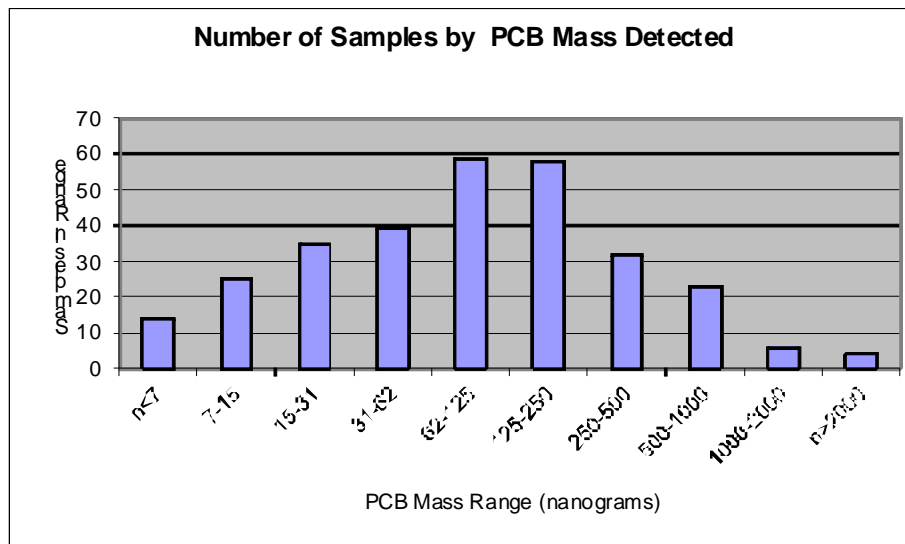


Figure 1. This bar chart shows the distribution of PCB masses detected on air sample cartridges used in the study. PCB air concentrations could not be calculated because the sample volumes were unknown.

The mean Cl/PCB ratio was 4.35 and the median value was 4.41. Approximately 20 air samples appeared dominated by an Aroclor 1260 source; these had Cl/PCB ratios of between 4.98 and 5.94. Approximately 180 samples appeared dominated by an Aroclor 1254 source; these had Cl/PCB ratios of between 4.00 and 5.27. Fewer than 10 air samples are dominated by Aroclors 1221 or 1232. The remaining air samples appear to be a combination of Aroclor mixtures containing 1016/1242, 1248 and/or 1254; the Cl/PCB ratio for these samples range from 4.20 to 2.46. The last column in Table 1 shows the Cl/PCB ratios for the common Aroclor PCB mixtures and Figure 2 shows the distribution of CL/PCB ratios for the study samples.

Few samples contained octa-chloro, nona-chloro or deca-chloro homologs. For those that did, it is suspected that particulate matter in the sample may have contributed to the PCB load.

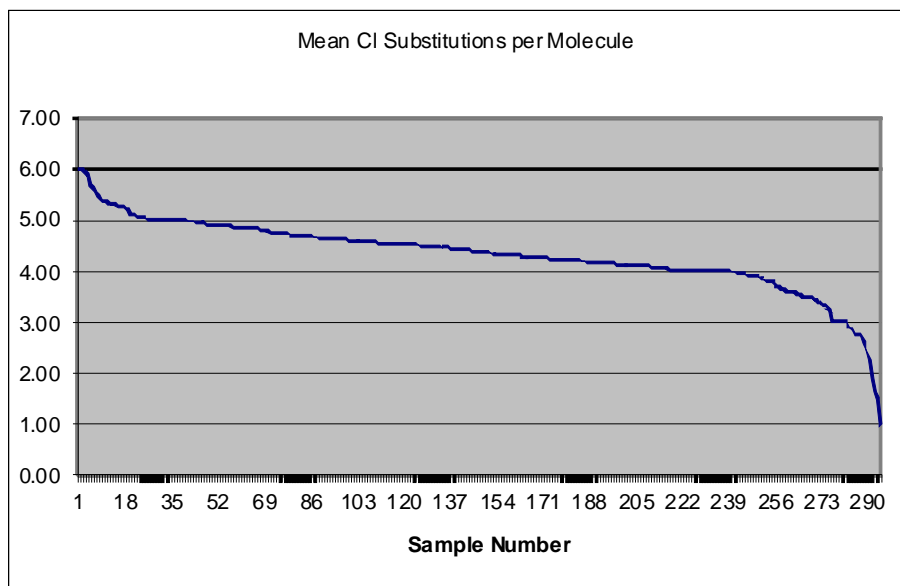


Figure 2. This chart shows the average chlorine substitutions per PCB molecule (along the y-axis) for the study sample set in descending order. Note one sample contained solely decachlorobiphenyl and that sample is not represented on the chart.

3.3 Comparison of Air Sample Homologs to Aroclors

For approximately two-thirds of the air samples, the Aroclor source of the PCBs was readily apparent from the homolog distribution in the air sample. However, even in these samples the homolog distribution was clearly shifted towards a greater proportion of lower chlorinated homologs relative to the parent Aroclor formulations. This shift towards lower chlorinated homologs is believed to be attributable to their overall greater volatility.

3.4 Variability of Aroclor 1254

Among the air sample set used in this study, Aroclor 1254 was by far the most frequently identified PCB mixture. In the course of identifying typical congener and homolog distribution patterns for the Aroclors, 1254 proved to be the most variable one in the literature. In his 1996 work, Frame discussed the variability he observed in Aroclor 1254 and noted that he had not observed nearly as much variability in the other Aroclor mixtures he analyzed.

Frame described chemical profiles for two different lots of Aroclor 1254. One lot he referred to as “A4”, obtained from AccuStandard of New Haven, CT. A second lot was referred to as “G4” was obtained from an old General Electric source. Frame concluded that the A4 material contained much higher concentrations of the coplanar PCB congeners than did the G4 material. Later reports by other researchers referred to an Aroclor 1254 mixture with properties similar to Frame’s A4 as either “Aroclor 1254 (Late)” or as “Type 2 Aroclor 1254” (AccuStandard, 2011). The more common Aroclor 1254 (Frame’s G4) is often referred to as “Type 1 Aroclor 1254”.

3.4.1 Type 1 Aroclor 1254

It is now believed that Type 1 Aroclor 1254 represents approximately 99% of the total Aroclor 1254 production, Type 2 made up the rest. Type 1 is believed to have been manufactured by a one-step chlorination process where biphenyl was chlorinated over a metal catalyst until it has achieved the desired degree of chlorination (54%). Type 1 Aroclor 1254 has generally been shown to have low concentrations of polychlorinated dibenzofurans (PCDFs) and coplanar PCBs (PCB 77, 126 and 169).

3.4.2 Type 2 Aroclor 1254

Type 2 Aroclor 1254 has a more complex and interesting history. It has its origins in Monsanto’s reaction to the 1968 mass poisoning incident in Japan, where PCB-containing heat transfer fluid leaked into rice bran oil intended for human consumption (Kuratsune et al, 2007). Thousands of people were seriously sickened in what became internationally known as the Yusho rice oil poisoning. The Yusho incident dramatically heightened concerns about the safety of PCBs (although subsequent studies found that it was actually other contaminants in the heat exchange fluid that caused the poisoning).

Monsanto decided to seek a safer alternative to the PCB formulations then on the market and in 1970-1971 they started selling Aroclor 1016. In its physical properties, Aroclor 1016 was equivalent to Aroclor 1242 (with 42% chlorine content), but it had much lower concentrations of the tetra-chloro and higher chlorinated homologs. Toxicity studies subsequently confirmed that Aroclor 1016 was less toxic than the other PCB formulations.

However, the process used to manufacture Aroclor 1016 resulted in a chlorinated biphenyl by-product that did not meet the Aroclor 1016 specification. These off-specification chlorinated biphenyls were subjected to a second chlorination step to bring that brought them up to 54% chlorine content. This “Type 2” Aroclor 1254 had the same physical properties and uses as Type 1, but it

had some distinctive chemical differences, including a significantly higher content of PCDFs and coplanar PCBs.

As sales of Aroclor 1016 increased, so did the production of Type 2 Aroclor 1254. It is not known whether the production of Type 2 Aroclor 1254 included the addition of “virgin” biphenyl being added to the Aroclor 1016 off-specification by-product. Sales and manufacturing of Aroclor 1254 ceased by 1977, but just prior to that time it is believed that Type 2 was the dominant Aroclor 1254 mixture being sold.

3.5 Aroclor 1254 Types in Building Materials

Since Type 2 Aroclor 1254 has a significantly higher content of the most toxic PCB constituents (the coplanar PCBs) in addition to higher concentrations of the highly toxic PCDFs, it is important to consider which type of Aroclor 1254 was used in building materials. At about the same time that Monsanto was developing Aroclor 1016, the company adopted its voluntary policy of not selling PCBs for uses that were not deemed to be “totally enclosed”. This was at about the same time that the first lots of Type 2 Aroclor 1254 were being produced. Therefore, it is likely that little if any Type 2 Aroclor 1254 was ever used in building materials, a use that is not "totally enclosed".

As a consequence of the general absence of Type 2 Aroclor 1254 from building materials it is reasonable to assume that the PCBs present in indoor air derived from building materials would be not be Type 2 Aroclor 1254.

3.6 Aroclor 1254 Types Used in Toxicological Studies

While there have been numerous toxicological studies conducted to assess the toxicity of PCBs, there are two principal studies that underpin EPA’s selection of the PCB toxicity factors (USEPA – IRIS, 2011). For the cancer slope factor (CSF), the 1998 Mayes et al (1998) (also cited as Brunner et al (1996)) was used. For the reference dose (RfD), the 1993 work by Arnold et al (1993) was the principal study.

The Mayes et al (1998) paper evaluated the toxicity of four different Aroclors in Sprague-Dawley rats. Among the four Aroclors studied, the Aroclor 1254 contained the highest concentrations of polychlorinated dibenzo-dioxins (PCDDs), PCDFs, and coplanar PCBs. This was identified by the authors as Type 2 Aroclor 1254. Prior to conducting the animal feeding study, the Aroclor 1254 was treated to eliminate the PCDFs, which also reduced the concentration of PCB 126 (the most toxic coplanar PCB) by 35%. The concentration of PCB 126 in the treated Aroclor 1254 was estimated to be 3 to 5 times greater than would be expected in typical Aroclor 1254 (Type 1). The authors estimated that the toxicity

equivalent factor (TEF) for their Aroclor 1254 was about twice the TEF for typical Aroclor 1254.

The Arnold et al (1993) work, which was used by the EPA to establish the PCB RfD, also included the results of Aroclor 1254 analysis for the lot used in his monkey feeding study. Citing the results of an unpublished memo, this lot of Aroclor 1254 was reported to contain no PCDD (<0.1 ppm); 5.2 ppm total PCDF; 0.05% PCB 77; 0.01% PCB 126; and 1 ppm PCB 169. It is not apparent from this information whether Arnold's lot of Aroclor 1254 is Type 1 or Type 2. The PCDF and coplanar PCB contents are elevated above the levels expected for Type 1, but not sufficiently elevated to permit easy classification as Type 2.

4. CONCLUSIONS

From the results of indoor air PCB homolog testing, the characteristics of the parent Aroclor mixture could be readily discerned approximately two-thirds of the time, even though the air samples displayed significant deviations from the parent Aroclor homolog distributions. In most air samples (>80%) there was a strong shift towards lower chlorinated homologs relative to the parent Aroclor formulation. The Aroclor identified most frequently in the air samples (in approximately 65% of the samples) was Aroclor 1254.

Aroclor 1254 is known to have a higher degree of chemical variation from lot to lot than other Aroclors. This is due in part to the two-step manufacturing method used by Monsanto in the last years of its production (between 1971-1977). This two-step method resulted in significantly higher concentrations of PCDFs and coplanar PCBs in the Type 2 Aroclor 1254 than occur in the more Typical Type 1 material. As a consequence of the higher concentrations of toxic PCDFs and coplanar PCBs, there is a reasonable likelihood that Type 2 Aroclor 1254 is more toxic than Type 1 Aroclor 1254. The authors are not aware of comparative animal studies having been conducted to test this hypothesis.

Because of Monsanto's voluntary ban on the use of PCBs in other than totally enclosed applications (beginning in 1970-1971), it is unlikely that Type 2 Aroclor 1254 was used in building materials. As a result, indoor air containing Aroclor 1254 from building materials is likely to be Type 1.

At the time researchers began conducting toxicological studies on PCBs, much or all of the Aroclor 1254 being produced was the potentially more toxic Type 2, as this was what was generally available to researchers. In evaluating the potential risks from exposures to the much more abundant Type 1 Aroclor 1254, toxicity studies conducted using Type 2 Aroclor 1254 may significantly overestimate actual risk.

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