THE RELATIONSHIP BETWEEN LEVELS OF PCBs AND PESTICIDES IN HUMAN HAIR AND BLOOD: PRELIMINARY RESULTS

Larisa Altshul[1], Adrian Covaci[2], Russ Hauser[1]

[1]Department of Environmental Health, Harvard School of Public Health, 665 Huntington Ave., Boston, MA 02115, USA.
[2]Toxicological Centre, University of Antwerp, Universiteitsplein 1, 2610 Wilrijk, Belgium

Introduction

The analysis of human hair as a biological measure of exposure to environmental contaminants has several advantages over the commonly used blood or adipose tissue sample. Hair collection is not invasive, the hair can contain relatively high levels of contaminants, and the hair specimen is relatively stable. There is a significant literature describing methods for analyzing hair for metals (especially methyl mercury and arsenic) and drugs [1]. However, there are many limitations in interpreting and using hair analysis in assessing exposure to environmental contaminants. There are no standardized, consistent methods for sample collection, preparation, and washing procedures. It is also difficult to distinguish between exogenous contaminants (deposited externally) and endogenous (incorporated internally from the hair follicles). There are currently no reference values or ranges for background levels of contaminants in hair or levels that are considered elevated. Information describing the analysis of organic contaminants in hair is especially limited. Analytical method for extraction of PCBs, DDT and HCH isomers from human hair was reported by Covaci and Schepens [2]. The comparison between selected PCBs and pesticides levels in hair and breast milk from the same individual was also reported. To the best of our knowledge, there are no other data describing the correlation between levels of organic contaminants in human hair and other body compartments, such as blood or other target tissues.

The objectives of this pilot study were: 1) to validate analytical method for detecting PCBs and chlorinated pesticides in human hair, 2) to evaluate the effect of washing hair on the levels of contaminants, and 3) to measure levels of organochlorines in hair and blood serum of 10 adult volunteers and 4) to determine if there are correlations between these levels in hair and blood.

Materials and Methods

Study participants

Ten adults (5 men and 5 women) were enrolled in this pilot study. All subjects signed an informed consent approved by the Harvard School of Public Health Human Subjects Committee. There is no standard protocol for collecting hair samples, so hair was collected by study participants during their routine hair cut, and there was no consistency in collection location. Because men and some of the women had short hair it was cut from all regions of the scalp. For women with long hair only the distal portion of hair was cut.

Hair washing

Hair samples were covered with 35 ml of hot water, sonicated for 30 minutes, and then dried with paper towel. For washing hair with shampoo, we placed hair in 40 ml screw cap vial, filled it with 35 ml of deionized water, added one drop of mild «Johnson & Johnson baby shampoo» (containing the

following ingredients: water, PEG-80 sorbitan laurate, cocamidopropyl betaine, sodium trideceth sulfate, glycerin, disodium lauroamphodiacetate, PEG-150 distearate, sodium laureth-13 carboxylate, fragrance, polyquaternium-10, Na_4EDTA, quaternium-15, citric acid, D&C Yellow no. 10 and D&C Orange no. 4) and vigorously shook the vial for 3 minutes. The washing solution was decanted (and saved for analysis), the hair was rinsed 5 times, each time with 30 ml of deionized water and the rinses were added to washing solution. For the hair washed with shampoo twice, the washing procedure was repeated by adding shampoo the second time before rinsing the hair.

Laboratory analysis

Blood serum and hair samples were analyzed for 57 individual PCB congeners and chlorinated pesticides. Details of hair extraction [2], serum analyses [3] and washing liquid extraction [4] were already reported. For all three matrices, the extract was cleaned-up using a chromatographic column packed with anhydrous sodium sulfate and 3 % deactivated silica gel. The sample extracts were analyzed by dual capillary HRGC/ECD and quantitated based on the response factor of each analyte relative to the internal standard, PCB 166. The average values obtained from both columns were reported for each target analyte, unless the difference between two results exceeded 20 %, in which case the lower value was reported. All final concentrations were reported after subtracting the amount of the analyte measured in the procedural blank.

Analytical Accuracy and Reproducibility

The mean (SD) for SPCBs in procedural blanks for hair was 0.79 (0.08) ng. The average recoveries for two surrogates, PCB 30 and PCB 112 added to hair samples were 73 (5) % and 82 (7) % and for washing liquid they were 86 (18) % and 86 (20) %, respectively. The mean percent recovery for PCB congeners added to eight hair matrix spike samples was 91 % (39%). Relatively high standard deviation for these measurements could be due to not homogeneous hair matrix. Analytical precision, expressed as mean (SD) coefficient of variation for 6 triplicate and 3 duplicate hair samples was 9 (8) for SPCBs, 9 (7) for p,p'-DDE, 20 (9) % for percent lipid, and 7(5) % and 3 (3) % for recoveries of two surrogates.

Results and Discussion

This pilot study demonstrated that the analytical method for analyzing organic contaminants in human hair is reliable and reproducible. The percent lipids and levels of organic contaminants in hair were higher than in serum. For the group of 10 adults, the mean (SD) percent lipid and levels for S PCB, p,p'-DDE and p,p'-DDT in serum and hair are presented in Table 1.

Table 1. Percent lipids and levels of organochlorines in human serum and hair (n=10)

Matrix	Lipids	ΣPCB		p,p'-DDE		p,p'-DDT	
	%	ng/g	ng/g fat*	ng/g	ng/g fat*	ng/g	ng/g fat*
Serum	0.52 (0.19)	2.7 (1.7)	558 (312)	2.9 (3.1)	600 (580)	0.11 (0.10)	24 (28)
Hair	1.90 (0.85)	61 (64)	2875 (1870)	12 (18)	570 (810)	6.4 (8.7)	300 (390)

***** Percent lipid for serum samples was available for 9 samples only.

Washing hair with shampoo decreased the levels of PCBs, pesticides and lipids by 27-29 % on average (Table 2). For more volatile congeners, such as #8 and #18, this decrease was even higher, up

to 48 % and 62 %, respectively. This pilot study did not find a strong or consistent correlation between the levels of organochlorines in hair and blood. The ratios of hair to serum levels are presented in Table 3.

To compare profiles of contaminants in hair and serum, we expressed concentrations (in ng/g fat) of selected PCBs as weight percent (contribution of each compound to their sum) and presented the mean values for both compartments (Figure 1). As can be seen from Figure 1, the percent of highly chlorinated and more persistent PCBs is higher in serum. With the exception of congener 74, the percent of less chlorinated and more volatile PCBs is higher in hair, this can be attributed to external gaseous or particulates exposure. This trend is especially evident for congeners, which are more rapidly metabolized in humans, such as PCB 52, 101, 110, 149. They were present at high concentrations even in the hair washed twice with shampoo. This may be a result of imperfect hair structure with various scratches and holes in the matrix, acting as adsorbing sites.

Table 2. Changes in organochlorines levels in hair after washing it once and twice with shampoo

IUPAC #	Hot water Mean (n=3) (ng/g)	1 x Shampoo Mean (n=3) (ng/g)	Loss after one shampoo (%)	2 x Shampoo Mean (n=3) (ng/g)	Loss after 2 x shampoo (%)
8	0.65	0.33	49	0.34	48
18	0.39	0.21	46	0.15	62
28	1.2	0.92	23	0.81	33
52	3.6	3.1	14	2.9	19
74	1.6	1.1	31	1.0	38
101	6.5	4.8	26	4.5	31
77/110	6.0	5.1	15	5.6	7
149	4.7	3.4	28	3.2	32
118	6.9	5.1	26	4.8	30
153	9.2	6.6	28	6.3	32
105/141	4.0	2.9	28	2.7	33
138	8.5	6.4	25	5.9	31
180	3.6	2.6	28	2.4	33
170	1.8	1.3	28	1.2	33
Σ PCBs	110	85	23	80	27
p,p'-DDE	80	63	21	60	25
p,p'-DDT	35	27	23	25	29
o,p'-DDT	22	17	23	15	32
% fat	3.1	2.3	26	2.2	29

* After one shampoo wash ** After two shampoo washes

If these compounds are exogenous, then the washing procedure was not effective in their elimination, thus making it problematic to make a clear distinction between exogenous and endogenous contamination. If these compounds are endogenous, then the elimination pathway is clearly different from what is found in the organs. The hair root is vascularised and therefore «in contact with blood» so contaminants may enter the hair shaft via the hair root. If the person has recent exposure to contaminants (or had it continually at low/background concentrations), these contaminants will be present in the hair in relatively higher abundance. In organs and fat (depot tissues) their concentrations will be lower, because they will accumulate only after being metabolized via the liver.

Conclusions

Although the analytical method for organochlorines in hair is reliable and reproducible, this pilot study has several limitations, including the lack of consistency in hair collection and in the time period

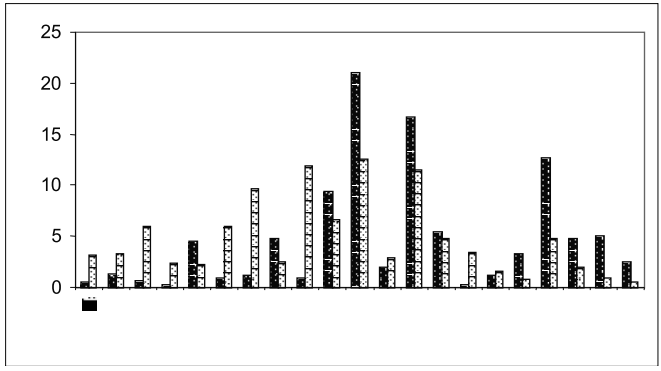

Figure 1. Profile of PCBs in human serum and hair

Table 3. Ratios of hair to serum concentrations (ng/g fat) for selected PCBs and pesticides

PCB	Women				Men				
IUPAC #	#1	#2	#3	#4	#1	#2	#3	#4	#5
28	3	5	15	3	8	9	12	21	8
52	5	26	170	12	38	18	95	76	19
74	1	2	7	0.3	0.3	0	1	3	2
101	20	8	174	6	41	46	34	105	16
149	46	13	115	13	62	89	26	146	19
118	2	1.7	8.2	0.3	4	2	4	6	1
153	2	1.3	4	0.3	3	2	3	5	1
105	10	1.2	16	0.3	5	3	43	5	4
138	3	1.5	6	0.4	2	3	4	6	1
180	1	1.5	1	0.3	2	1	3	2	1
170	1	1.7	1.2	0.3	2	1	4	3	1
Σ PCBs	3	3.1	15	1.0	6	4	7	12	3
p,p'-DDE	0.3	1.5	1.3	0.2	0.8	1	1	1	0.4
p,p'-DDT	2	28	35	2	2	18	13	5	1
o,p'-DDT	4	70	112	5	9	9	16	10	1

between collecting hair and blood samples from the same individual. More research is needed to establish if hair analysis could be useful and if it could be considered preferable in certain situations, such as for pediatric exposures.

Acknowledgements

The study was partially supported by NIEHS grants 5P42 ES05947, ES000002, and ES09718.

The authors are grateful to R. Stolyar and S. Forsberg for chemical analysis and to the volunteers participating in the study.

References

1. Morton J, Carolan VA, Gardiner PHE (2002) Anal Chim Acta, 455, 23-34.
2. Covaci A, Schepens P (2001) Chromatographia 53, S366-S371.
3. Korrick SA, Altshul LM, Tolbert PE, Burse VW, Needham LL, Monson RR (2000) J. Exp Anal Environ Epidemiol 10, 743-754.
4. US EPA (1984) Federal Register, 40 CFR Part 136, 49, No.209:89-104.