Gevao *et al. J Environ Expo Assess* 2022;1:5
DOI: 10.20517/jeea.2021.06

Journal of Environmental Exposure Assessment

Research Article

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Persistent organic pollutants on human and sheep hair and comparison with POPs in indoor and outdoor air

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How to cite this article: Gevao B, Uddin S, Al-Bahloul M, Al-Mutairi A. Persistent organic pollutants on human and sheep hair and comparison with POPs in indoor and outdoor air. *J Environ Expo Assess* 2022;1:5. https://dx.doi.org/10.20517/jeea.2021.06

Received: 2 Nov 2021 **First Decision:** 25 Nov 2021 **Revised:** 17 Dec 2021 **Accepted:** 11 Jan 2022 **Published:** 20 Jan 2022

Academic Editor: Stuart Harrad **Copy Editor:** Xi-Jun Chen **Production Editor:** Xi-Jun Chen

Abstract

This study compared the concentration of persistent organic pollutants (POPs) in air derived from polyurethane-based passive samplers to those of hair samples collected from humans and sheep. Human scalp hair samples were obtained from 24 healthy individuals and ten sheep (*Ovis aries*) during indoor and outdoor polyurethane foam plug ambient sampling. The samples were analyzed for polycyclic aromatic hydrocarbons (PAHs) and polybrominated diphenyl ethers (PBDEs). ∑PBDE concentrations ranged 0.6-50 ng·g^{-1} (mean, 18.6 ± 13 ng·g^{-1}) for humans and 0.6-1.4 ng·g^{-1} (mean, 1.1 ± 0.25 ng·g^{-1}) for sheep. The ∑PAH concentrations were log-normally distributed in human hair ranging 98-2529 ng·g^{-1} (mean, 460 ± 538 ng·g^{-1}), whereas concentrations for sheep hair samples ranged 168-526 ng·g^{-1} (mean, 334 ± 117 ng·g^{-1}). Strong correlations (*P*-values < 0.01) were found between concentrations of PAHs and PBDEs in human and sheep hair with concentrations measured in indoor and outdoor air, respectively. Evidence generated from this preliminary study suggests that hair might be used for the environmental monitoring of POPs in remote sites to provide a first-order estimate of ambient levels. Further studies are required to understand the uptake profiles and validate the use of hair as a sampling medium for POPs in ambient air.

Keywords: Indoor air, outdoor air, hair, PBDE, PAHs

INTRODUCTION

Persistent organic compounds such as polycyclic aromatic hydrocarbons (PAHs) and polybrominated diphenyl ethers (PBDEs) have the propensity to enter the gas phase at ambient temperatures and undergo long-range atmospheric transport. These chemicals have received intense international attention because of their ubiquity, bioaccumulation potential, and detrimental biological effects[1-3]. The combination of their resistance to metabolism and lipophilicity means that they will bioaccumulate and be transported through food chains. In addition, they are subject to long-range transport and have been detected in remote areas where they have never been used[4,5]. Since atmospheric transport is the principal vehicle for the movement and global distribution of these chemicals, significant efforts are being made on identifying their ambient sources[6-9], transport pathways[4,10,11], and fate[12-14]. Until recently, there was a paucity of reliable environmental data on the levels of most POP chemicals in the Middle East, most of Africa, and Asia, from which to assess the effectiveness of international efforts to minimize the release of these chemicals to the environment. This is partly due to the lack of appropriate analytical facilities and trained personnel in developing countries. High-volume air sampling procedures, for example, are expensive, often require skilled personnel for sampling, and require electricity for their operation. Recently, passive samplers such as semipermeable membrane devices[15,16], polyurethane foam (PUF)[17-19], polymer-coated glass[20,21], XAD filled tubes[22], and tristearin-coated fiberglass sheets[23] have been developed and validated for measuring POPs in the atmosphere.

It is desirable to have alternative sampling technologies that will provide identical information at lower costs. In the past, vegetation such as grass, pine needles[24-26], mosses[27,28], and lichens[29] were studied to make inferences about POP concentrations in the air. The main advantage of passive sampling is that it is cheap and does not require skilled personnel for its deployment, which makes widespread deployment possible especially in remote locations. The simplistic design and operation have made passive samplers attractive for developing countries that are still developing their capabilities of POPs sampling and analyses. Some studies have used hair as a potential matrix for estimating semi-volatile organic pollutants in air[30,31] and for POP assessment[32-35]. Since hair has a high lipid content, it can be a suitable medium for the retention of POPs. Hair has similar attributes with other passive samplers, including the simplicity of sample collection, storage, and transportation[31,36-38]. Sample handling protocols are least stringent for hair, and, as such, sampling does not require very skilled personnel. Since hair sampling is non-invasive, subject compliance is high as it is socially and ethically acceptable compared to other samplers. Samples can be easily obtained from people of different age groups and sex. As such, broader geographical coverage can be achieved with little effort[31,36-38], making large-scale mapping exercises and reconnaissance surveys very feasible and cost-effective.

Some studies have tried to investigate the link between concentrations of POPs in hair and internal tissues in humans and animals to determine whether hair concentrations reflect body burdens[31,36-41]. Tirler *et al.*[42] (2001) suggested that hair could be used as a passive sampler for POPs when determining the indoor air levels of lindane in rural Germany. The most direct evidence, so far, to assess the potential of hair as a passive sampler of POPs comes from the work of Schramm[30]. In laboratory microcosm experiments, hair was fumigated with gaseous PCBs and PCDD/Fs, and the kinetics of uptake were evaluated. Rapid uptake was observed, with equilibrium established within hours of exposure. The current study investigated the comparability of POP concentration in air derived from polyurethane-based passive sampling and hair to assess if hair can be used for the environmental monitoring of POPs. The approach we adopted was to

passively sample the air in homes of volunteers that consented to provide hair samples, using PUF passive samplers deployed over a six-week period. In addition, hair was sampled from ten randomly selected sheep during the same period as the outdoor deployment of passive samplers. Indoor air and ambient (outdoor) air concentrations have been published previously[17,43]. Here, we report the concentrations of PAHs and PBDEs in hair and examine the relationships between hair concentrations in air and passive samplers.

EXPERIMENTAL

Sample collection and pretreatment

Hair samples

Twenty-four healthy males in a broad age group were selected for the collection of scalp hair samples in Kuwait. Passive samplers were deployed in their homes. Using stainless steel scissors, sheep (*Ovis aries*) hair samples were collected across Kuwait from 10 randomly selected sheep. The hair samples were stored at -20 °C before sample preparation in solvent rinsed, clean amber glass jars. Hair samples were washed by sonication in an ultrasonic bath using 35 mL of water for 2 h to remove associated dust. They were then dried with paper towels and ground into a fine powder using a grinding mill (Retsch, Germany). The finely ground hair was stored at -20 °C before chemical analysis.

Passive samples

The PUF disks used to collect chemicals from the air were certified as flame retardant free and purchased from Tisch Environmental (OH, USA). The PUF disks were cleaned for 48 h using dichloromethane in a giant Soxhlet. The pre-extracted PUF plugs were dried in a clean desiccator under vacuum and stored in solvent rinsed amber glass jars lined with solvent rinsed aluminum foil to avoid contamination during storage. In the field, the PUF was suspended in the center of two stainless steel dishes between washers by using solvent-rinsed tweezers. The samplers were attached to a pole at the site of deployment. Outdoor deployment was on roof-tops where the air could be considered well mixed. Samples were deployed at 17 sites over a six-week period.

Chemicals and reagents

Analytical grade solvents were procured from VWR Scientific (NY, USA). The silica (100-200 mesh), alumina, and sodium sulfate used were manufactured by Baker (NJ, USA). The deuterated PAH cocktail standard ES-2044 was used as the internal standard. This standard contains pyrene-d_{10}, phenanthrene-d_{10}, fluoranthene-d_{10}, benzo[a]pyrene-d_{12}, and benzo[*ghi*]perylene-d_{12}. The other analytical standard used was EO-5103 for PBDEs, with a congener mix of the following: 17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190, and 209. Additionally, brominated diphenyl ether (BDE) 35 (EO-4109) and BDE 181 (EO-4927) were purchased from Cambridge Isotope Laboratories (CIL, Andover MA, USA) were added at a concentration of 100 ng.

Extraction and analyses

PUF disk samples were extracted in a Soxhlet apparatus using a 1:1 *v/v* mixture of acetone:hexane. Prior to extraction, the samples were spiked with a range of surrogates to monitor analytical recovery. The extracts were cleaned by column chromatography using 10 g of silica and 5 g of alumina column. They were further cleaned by gel permeation chromatography using 6 g of Biobeads SX 3 (BioRad, Hertfordshire, UK). The final volume was made up with 500 μL of isooctane and spiked with internal standards.

For the extraction of hair samples, 0.2-0.5 g of finely ground hair were weighed in a 40 mL vial; spiked with a range of ES-2044, EO-5103, EO-4109, and EO-4927; and incubated overnight at 40 °C in 4 mL of a 3 N HCl and 3 mL of hexane:dichloromethane (4:1, *v/v*) to monitor analytical recovery. The analytes of interest

were extracted from the incubation medium by liquid-liquid extraction with 2 × 4 mL hexane:dichloromethane (4:1, *v/v*). The organic extracts were combined and dried on a bed of anhydrous Na_2SO_4 to remove any residual water. Column chromatography was used for removing the interfering compounds, using 2 g of silica and 1 g of alumina (and 0.5 cm anhydrous Na_2SO_4 at the top of the column to prevent the column from coming into contact with air) and eluting the compounds of interest with 40 mL 1:1 mixture of hexane:DCM. The eluent was blown down under a gentle stream of nitrogen. The final volume was made up of 500 μL of isooctane and spiked with mirex (10 μL of 10 ng/μL) as an internal standard.

The PAHs in the sample extracts were analyzed using splitless injection (injection volume, 1 μL) on a 30 m HP-5ms column (0.25 mm i.d., 0.25 μm film thickness) on a Shimadzu GC-17A (Shimadzu, Tokyo, Japan) gas chromatograph using helium as a carrier gas. The method is described in detail elsewhere[44]. An internal standard method for identification and quantification against five calibration standards was used that contained fifteen PAHs (acenaphthylene, acenaphthene, anthracene, benz[*a*]anthracene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, benzo[*ghi*]perylene, chrysene, dibenz[*a,h*]anthracene, fluorene, fluoranthene, indo[1,2,3-*cd*]pyrene, phenanthrene, and pyrene), although not all were routinely detected in samples. The sum of the concentrations of these PAHs was designated as ∑PAHs.

Consequent to the PAH analyses, the sample volume was reduced to 50 μL in dodecane for PBDE analysis. PBDEs were analyzed using splitless injection on a 30 m HP-5ms column (0.25 mm i.d., 0.25 μm film thickness) with an Agilent 6890N gas chromatograph using helium as carrier gas. Details of the method are given elsewhere[45]. Identification and quantification were carried out against five calibration standards. BDE 209 was analyzed on a 15 m DB-5ms column (0.25 mm i.d., 0.25 μm film thickness). The GC oven conditions, identification, and quantitation followed those of Gevao *et al.*[45].

QA/QC

Peaks were positively identified if they were within ± 0.05 min of the retention time in the calibration standard. They were quantified only if the response exceeded three times the background noise, and the isotopic ratio between the quantitative and the confirmation ions was within ± 20% of the theoretical value[45]. The concentration of fortified hair samples was extrapolated to a signal-to-noise level of 3 for calculating the detection limit[46]. The detection limits for PBDEs ranged 5-60 $pg \cdot g^{-1}$ with the exception to BDE 209 for which the detection limit was 1.5 $ng \cdot g^{-1}$. The detection limits for PAHs ranged 1.0 ≠ 12.8 $pg \cdot g^{-1}$. Laboratory blanks, comprised of incubation solution and treated as samples, were processed for every five samples. The concentration in blanks was subtracted from those in the sample extracts. Average recoveries [(%) ± standard deviation (SD)] for surrogates spiked in samples were between 85% ± 10% for BDE 35, 74% ± 5% for BDE 181, whereas those of PAHs varied from 65% ± 8% to 95% ± 5%.

RESULTS AND DISCUSSION

The estimated air concentrations were derived assuming a sampling rate of 3.0 m^3 air per day[47,48] derived from calibration studies against an active sampler. Applying this sampling rate, the PUF disks would have sampled 105 m^3 of air over the six-week deployment period. The sampling rate has been shown to be linear for compounds with K_{OA} larger than $10^{8.5}$ for the first 100 days.

PBDEs in hair

The following congeners were detected: BDEs 28, 47, 99, 100, 153, 154, 183, and 209. Their sum is referred to as ∑PBDEs. None of the sheep samples contained BDEs 183 or 209 above detection limits. ∑PBDE

concentrations measured in human and sheep hair samples are summarized in Figures 1 and 2, respectively, and detailed congener-specific measured concentrations are provided in Tables 1 and 2, respectively. Concentrations ranged 0.6-50 ng·g^{-1} of human hair with a mean of 18.6 ± 13 ng·g^{-1}. For ∑PBDEs, in human hair, BDE 209, constituted ~87%, followed by BDE 47 (7.3%), BDE 99 (7.1%), and BDE 28 (2.8%) with the remaining congeners (BDEs 100, 153, and 154) together contributing approximately 3.5%. In sheep hair samples, the concentrations of ∑PBDEs ranged 0.609-1.410 ng·g^{-1} with a mean of 1.100 ± 0.250 ng·g^{-1}. BDE 47 was the most abundant congener with ~54% of ∑PBDEs. This was followed by BDE 99 (27%), BDE 100 (7.5%), BDE 28 (6%), and BDEs 153-154 (2.4%).

The concentrations in human hair very closely track indoor air concentrations [Figure 3A], and the two are remarkably correlated (*P*-value = 0.004). Similarly, a significant correlation (*P*-value < 0.001) is observed between sheep hair and outdoor air concentrations of PBDEs estimated from passive sampling measurements across Kuwait [Figure 3B][17]. Other studies have reported PBDEs in animal hair; for example, D'Havé *et al.*[37] studied the concentrations in hair and other body tissues in 32 European hedgehogs (*Erinaceus europaeus*). Concentrations in hair were reported to vary between 0.8 and 11 ng·g^{-1} with a mean of 9 ng·g^{-1}. Recently, Jasper *et al.*[31] reported ∑PBDEs (sum of 47, 99, and 153) in the feathers of buzzards (*Buteo buteo*), a predatory bird, to vary from 0.5 to 10 ng·g^{-1} with a mean of 1.4 ng·g^{-1}.

PBDE congener profiles

As mentioned above, the congener distribution of human hair is dominated by BDE 209, which constitutes ca. 87% of the ∑PBDEs measured in human hair. This congener is, however, absent in sheep hair samples. In a previous study, high levels of BDE 209 were found in house dust in Kuwait[49]. The dominance of BDE 209 in dust and human hair samples suggests that deca-technical mixture is a dominant contributor of PBDEs to indoor air in Kuwait. If we exclude BDE 209 and express the congener contribution as the percentage of BDEs 28, 47, 100, 99, 85, 153, 154, and 183, it shows that the penta-congener mixture is an important technical mixture in Kuwait. The congener distributions in hair, indoor air, and outdoor air, excluding BDE 209, as well as of Bromkal 70-5DE, a commercially marketed mixture for comparison[50], are given in Figure 4. There are a few observations of importance in this profile. First, the composition in human hair very closely matches the technical penta-mixture, suggesting volatilization from penta-treated products is a significant indoor source. Second, the proportion of BDE 47, the dominant congener in the technical mixture in sheep and ambient air, is a reflection of the differences in the volatility of the congeners relative to each other. This hypothesis comes from the work by Bruckman *et al.*[51], who reported a five-fold increase in PBDE levels in a room after the TV was left on for several hours. In another study, volatilization of BFRs from television, computers, and printers was reported[52]. The authors concluded that annual BFRs emissions from TV and monitors were about 0.1% and 0.4%, respectively.

PAHs concentrations in hair

The concentration of PAHs in human hair is summarized in Figure 5, and detailed compound-specific information is given in Figure 6A and Table 3. The concentration of ∑PAHs varied from 98 to 2529 ng·g^{-1} of hair with a mean concentration of 460 ± 538 ng·g^{-1}, dominated by low molecular weight tricyclic PAHs contributing 75%. Phenanthrene was the most abundant compound contributing ca. 42%, followed by fluorene (12%), anthracene-fluoranthene (10%), and pyrene (7%). In the only other study of PAHs in human hair, Toriba *et al.*[53] reported a mean ∑PAH concentration of 176 ng·g^{-1}, which is lower than those one in this study by a factor of ~3. The compound distribution in their study is similar to that reported here, with the major compounds in decreasing order of importance being phenanthrene (60%), fluoranthene (14%), pyrene (10%), and fluorene-anthracene (5%). The high molecular weight PAHs were below the detection limits, as is the case in this study.

Table 1. PBDE concentrations (ng·g^{-1}) in human hair

	Congeners								
	28	**47**	**100**	**99**	**153**	**154**	**183**	**209**	**∑PBDEs**
H 1	0.30	3.23	0.42	1.42	0.20	0.08	0.26	40.61	46.52
H 2	0.09	1.25	0.21	1.32	0.09	0.14	0.05	27.04	30.19
H 3	0.04	0.79	0.12	0.82	0.07	0.02	0.10	14.73	16.68
H 4	0.07	1.60	0.36	1.43	0.09	0.14	0.03	16.11	19.83
H 5	0.04	0.99	0.12	1.41	0.03	0.13	0.05	14.76	17.53
H 6	4.84	5.61	1.51	2.59	0.46	1.35	0.30	16.22	32.88
H 7	0.07	1.72	0.16	1.21	0.05	0.07	0.04	8.75	12.06
H 8	4.38	4.96	1.17	3.04	0.41	1.18	0.25	15.10	30.49
H 9	0.05	0.71	0.09	0.78	0.05	0.11	0.25	9.43	11.46
H 10	0.04	1.11	0.12	0.15	0.04	0.03	< d.l.	13.02	14.52
H 11	0.12	0.97	0.48	0.56	0.03	0.06	0.09	24.23	26.55
H 12	0.02	0.27	0.04	0.18	0.01	0.02	0.04	49.59	50.18
H 13	0.03	0.70	0.12	0.86	0.06	0.11	0.05	8.18	10.11
H 15	0.06	0.96	0.17	1.32	0.07	0.12	0.06	12.00	14.76
H 16	0.03	0.67	0.14	0.11	0.04	0.11	< d.l.	< d.l.	1.10
H 17	0.07	0.36	0.06	0.08	0.00	0.03	0.02	< d.l.	0.63
H 18	0.01	0.07	0.22	0.27	0.00	0.01	0.02	5.23	5.83
H 19	0.01	0.12	0.02	0.12	0.02	0.02	0.01	2.17	2.49
H 20	0.07	0.89	0.09	0.57	0.05	0.08	0.03	13.42	15.20
H 21	0.05	1.55	0.08	9.96	0.04	0.08	0.30	12.12	24.18
H 22	0.04	1.41	0.25	0.06	0.07	0.28	0.09	20.19	22.39
H 23	0.04	0.50	0.09	0.72	0.03	0.05	0.07	11.05	12.55
H 24	0.02	0.45	0.07	0.47	0.03	0.06	0.08	9.08	10.25

PBDEs: Polybrominated diphenyl ethers.

Table 2. PBDEs in animals (pg·g^{-1})

	Congeners								
	28	**47**	**100**	**99**	**153**	**154**	**183**	**209**	**∑PBDEs**
AH 1	77.3	631.5	55.8	255.5	14.5	12.4	< d.l.	< d.l.	1047.0
AH 2	38.8	350.8	47.8	264.7	24.8	19.2	< d.l.	< d.l.	746.1
AH 3	89.7	801.6	78.0	362.2	35.9	42.4	< d.l.	< d.l.	1409.8
AH 4	61.5	475.3	59.5	297.8	36.9	21.6	< d.l.	< d.l.	952.6
AH 5	73.7	666.0	76.4	298.7	17.9	23.5	< d.l.	< d.l.	1156.3
AH 6	64.4	542.0	50.8	290.1	22.9	45.9	< d.l.	< d.l.	1016.2
AH 7	69.7	786.8	93.6	409.6	14.9	18.5	< d.l.	< d.l.	1393.1
AH 8	73.0	659.3	66.8	331.6	25.5	32.6	< d.l.	< d.l.	1188.9
AH 9	62.6	640.5	66.4	295.4	26.0	12.4	< d.l.	< d.l.	1103.3
AH 10	27.5	237.0	201.4	115.7	27.0	< d.l.	< d.l.	< d.l.	608.6
Mean	63.8	579.1	79.7	292.1	24.6	25.4	< d.l.	< d.l.	1062.2

PBDEs: Polybrominated diphenyl ethers.

Average ∑PAH concentrations in indoor air, concurrently measured using PUF samplers at the time of hair sampling, varied between 1.3 and 16 ng·m^{-3}[17]. The compositional profile of ∑PAHs in human hair measured in this study matches closely that of the indoor air profile (P-value < 0.001) [Figure 6A]. Concurrently measured air concentrations obtained by deploying PUF samplers across Kuwait ranged

Table 3. PAHs in human hair (ng·g^{-1})

	Compounds											
	Ace	**Acy**	**Fluo**	**Phen**	**Anthr**	**Fla**	**Pyr**	**B[a]A**	**Chry**	**B[b]F**	**B[k]F**	**∑PAHs**
H 1	40.3	265.6	217.9	537.7	20.4	72.8	74.8	3.7	10.5	< d.l.	< d.l.	1244
H 2	15.3	66.9	76.8	219.6	7.6	29.4	36.0	1.2	4.0	< d.l.	< d.l.	457
H 3	8.1	41.4	39.1	142.4	5.7	30.0	29.7	2.6	6.6	< d.l.	< d.l.	306
H 4	2.1	36.9	35.0	130.7	12.7	20.5	33.8	1.9	7.3	< d.l.	< d.l.	281
H 5	3.0	45.8	111.2	235.6	21.4	22.0	28.7	1.6	3.1	11.7	14.3	498
H 6	57.2	17.0	14.7	74.0	5.2	21.9	29.5	3.3	9.9	< d.l.	< d.l.	233
H 7	6.6	48.7	40.0	103.4	3.7	13.7	17.4	0.7	2.7	< d.l.	< d.l.	237
H 8	24.7	24.2	18.1	68.2	2.9	17.6	23.1	2.4	6.7	249.0	200.1	637
H 9	0.9	13.9	17.0	95.8	7.1	32.2	28.0	2.3	7.0	< d.l.	< d.l.	204
H 10	2.3	20.5	25.2	71.1	4.3	10.1	18.4	1.5	3.5	< d.l.	< d.l.	157
H 11	64.2	34.2	39.4	138.2	10.5	24.3	39.7	3.2	6.7	51.9	< d.l.	412
H 12	31.1	9.0	9.5	44.0	2.5	9.1	14.3	4.6	3.9	< d.l.	< d.l.	128
H 13	3.0	15.0	47.1	238.2	24.8	15.4	18.6	1.0	2.1	< d.l.	< d.l.	365
H 15	14.9	32.6	124.3	552	125.4	153.3	99.5	4.7	8.0	< d.l.	< d.l.	1114
H 16	2.4	10.3	14.4	54.2	4.2	9.6	12.9	1.7	3.3	< d.l.	< d.l.	113
H 17	25.5	35.8	443.0	537.6	852.6	537.7	232.0	2.2	6.3	< d.l.	< d.l.	2529
H 18	0.3	1.8	21.0	161.4	16.8	7.8	7.9	0.3	0.9	< d.l.	< d.l.	218
H 19	0.2	4.1	22.7	392.8	56.0	42.9	22.5	< d.l.	1.4	< d.l.	< d.l.	543
H 20	6.3	40.7	45.4	130.6	6.0	18.1	18.7	1.2	2.8	< d.l.	< d.l.	270
H 21	0.9	15.5	15.6	45.1	1.5	7.7	8.0	0.7	3.0	< d.l.	< d.l.	98
H 22	1.6	27.2	25.3	69.6	2.8	9.0	9.5	0.9	1.5	< d.l.	< d.l.	147
H 23	5.9	42.3	38.7	104.6	4.0	13.8	11.4	0.8	2.1	< d.l.	< d.l.	224
H 24	2.7	22.3	26.8	81.7	4.1	16.4	16.2	1.5	4.4	< d.l.	< d.l.	176

PAH: Polycyclic aromatic hydrocarbon.

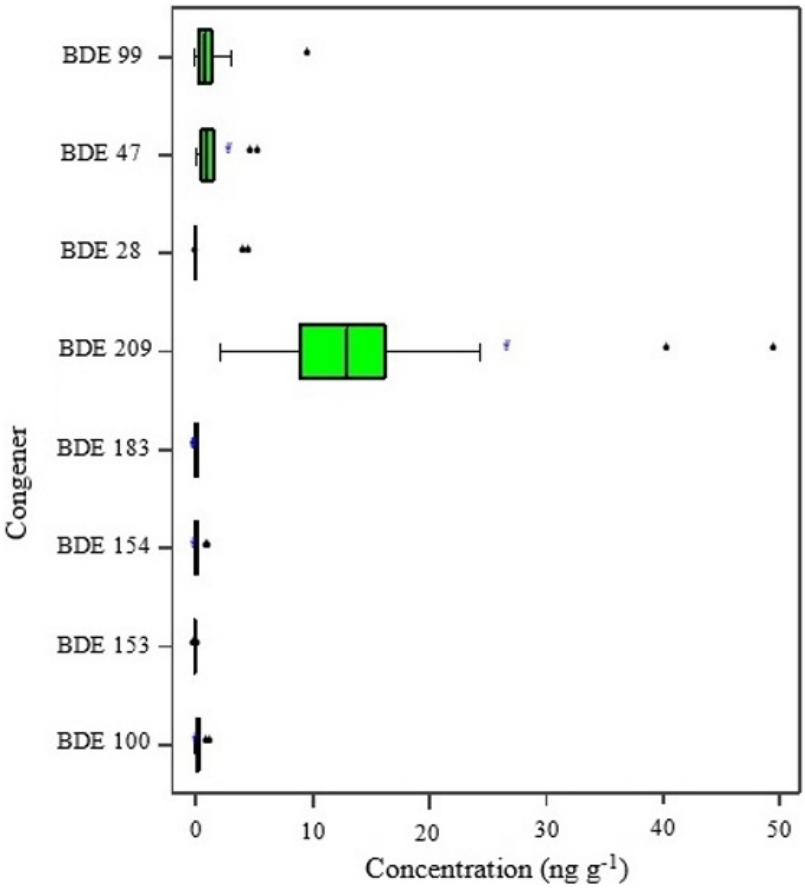

Figure 1. ∑PBDE concentrations (ng·g^{-1}) in human hair. PBDE: Polybrominated diphenyl ether.

between 5 and 13 ng·m^{-3}[17] during the same period sheep hair samples were obtained. The average concentrations of individual compounds in sheep hair closely track ambient air concentrations [Figure 6B] and are significantly correlated (P-value < 0.001).

In sheep hair samples, ∑PAH concentrations varied from 168 to 526 ng·g^{-1} [Figure 7] with a mean concentration of 334 ± 117 ng·g^{-1}. Detailed compound-specific information is given in Figure 6B and Table 4. The contributions of tricyclic PAHs and tetracyclic PAH were 74% and ca. 22% of the ∑PAHs. The major compounds in order of importance were phenanthrene (44%), fluorene (16%), anthracene (11%), fluoranthene (10%), and pyrene (9%).

Hair *vs.* passive air samplers

The vast majority of studies on pollutants in hair have focused on investigating the relationship between concentrations of pollutants in hair and other body tissues with the view to determine if hair can be used to estimate the contaminant body burden of humans and animals. These studies have presented consistent evidence in support of this hypothesis. Jaspers *et al.*[31], for example, reported significant correlations between feathers of *Buteo buteo*, a predatory bird, and other body tissues for some PBDE congeners, but low ones for DDT and others. They suggested this mixed result was due to differences in metabolic rates or differences in external contamination of the feathers. D'Havé *et al.*[37], also studying POPs in mammals, reported correlations between hair and body tissues (liver, kidney, and muscle) with coefficients varying between 0.72 and 0.78. A lack of any correlation in the case of BDE 99 was also attributed to differences in

Table 4. PAH concentrations (ng·g^{-1}) in animal hair

	AH 1	AH 2	AH 3	AH 4	AH 5	AH 6	AH 7	AH 8	AH 9	AH 10
Ace	16.0	15.6	11.1	15.9	0.9	19.4	25.5	5.7	15.9	10.6
Acy	23.7	20.3	23.1	22.2	1.8	32.5	36.7	23.3	34.7	23.0
Flu	90.7	63.3	100.7	83.8	10.5	117.6	135.3	103.7	125.8	57.4
Ph	281.6	160.7	345.7	210.6	139.9	321.2	334.7	360.4	325.4	143.5
An	14.4	6.8	16.4	10.3	6.4	16.8	9.6	21.0	4.7	8.5
Fla	47.9	28.7	77.6	40.4	48.8	50.1	60.7	73.1	56.4	42.5
Py	41.3	27.4	70.9	74.3	39.3	43.5	51.2	64.6	37.5	41.4
B[a]A	3.9	2.9	10.8	2.9	4.0	4.6	6.2	6.2	3.4	6.5
Chry	8.8	6.0	34.9	9.1	9.8	9.2	14.7	14.8	13.4	15.5
BbF	12.6	5.7	17.0	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	9.5
BkF	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.
BaP	3.1	4.1	6.1	1.8	2.6	2.8	3.9	3.7	2.1	2.9
I[123-cd]P	4.2	3.0	13.0	3.8	3.0	3.0	5.5	5.2	3.8	7.3
DahA	5.0	< d.l.	1.2	2.9	1.1	1.5	1.2	1.7	1.8	3.6
BghiP	5.4	4.2	17.1	13.6	3.6	3.8	5.9	8.3	5.1	9.1
∑PAHs	**558.7**	**348.5**	**745.6**	**491.5**	**271.8**	**626.1**	**691.0**	**691.7**	**630.1**	**381.1**

PAHs: Polycyclic aromatic hydrocarbons.

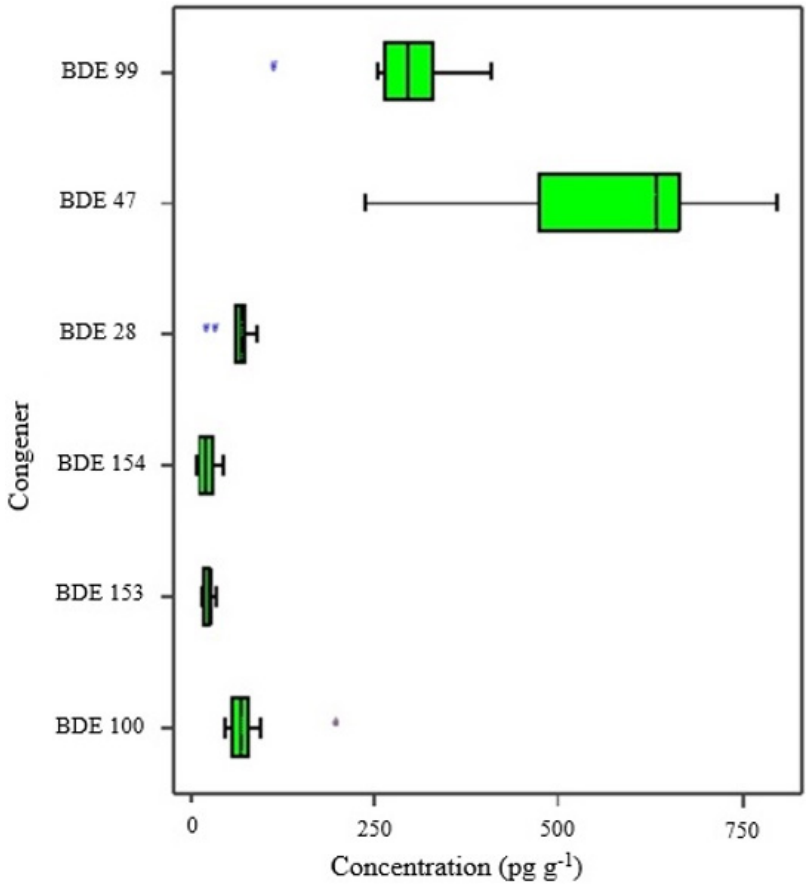

Figure 2. ∑PBDE concentrations (pg·g^{-1}) in Sheep hair. PBDE: Polybrominated diphenyl ether.

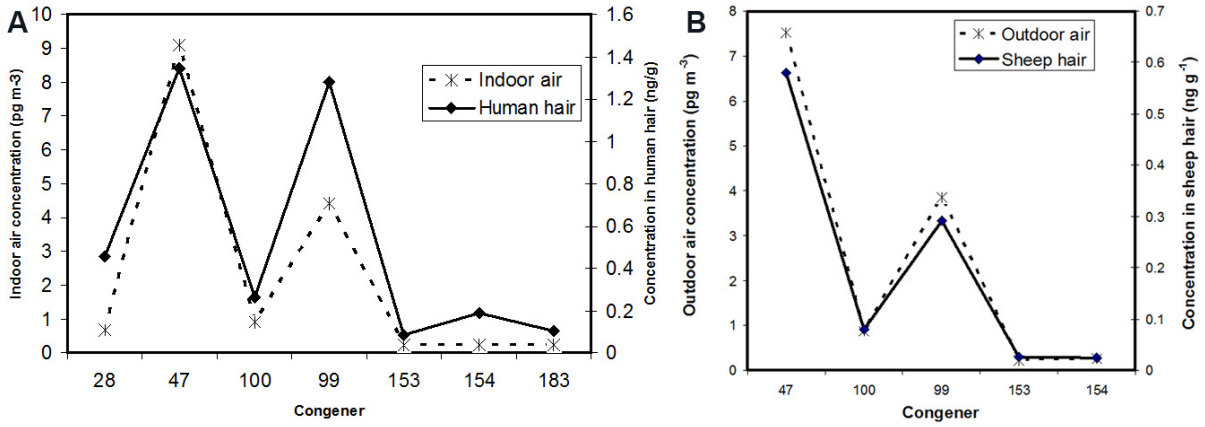

Figure 3. (A) The PBDE congener profile in indoor air and human hair samples (ng/g). (B) The congener profile of PBDEs in outdoor air and the sheep hair samples. PBDEs: Polybrominated diphenyl ethers.

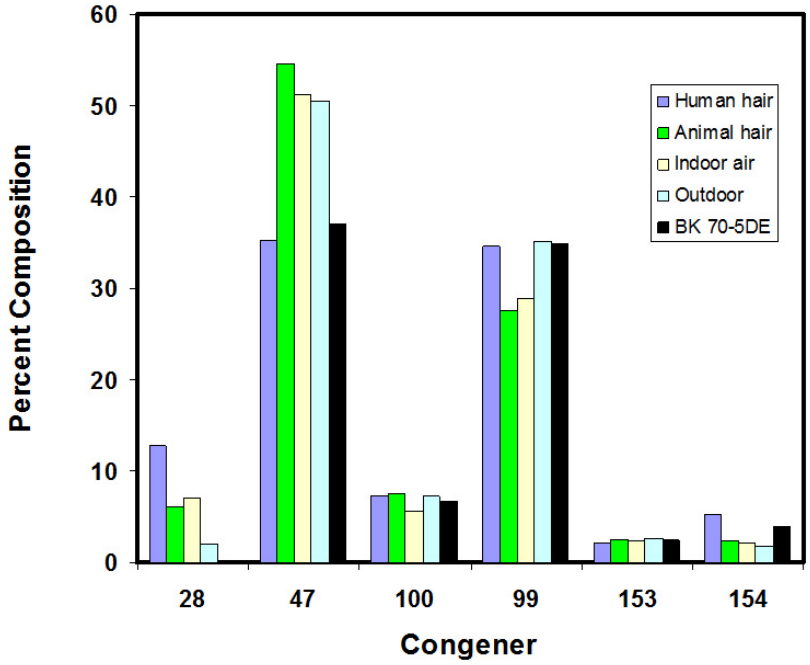

Figure 4. The percent distributions of ∑PBDE congeners in hair, indoor air, outdoor air, and Bromkal 70-5DE. PBDEs: Polybrominated diphenyl ethers.

accumulation rates between different tissues and hair. Gill *et al.*[54] reported relatively higher concentrations of PCB 52 and 101 in hair compared with those in adipose tissues or serum. The authors contended that this was due to contamination of hair from exogenous sources and/or possible differences in elimination kinetics between congeners. Altshul *et al.*[38] reported a significant correlation (r = 0.8) between hair and the serum of 10 individuals for *p,p′*-DDE but non-significant correlations for PCBs and *p,p′*-DDT. Covaci and Schepens[39] reported similar profiles of PCBs in human hair, serum, milk, and adipose tissue. When concentrations were normalized to lipid, similar concentrations were found among all the matrices for lindane, PCBs, *p,p′*-DDT, and *p,p′*-DDE. Nakao *et al.*[55] reported similar ratios of PCDD/Fs in the blood and hair of six donors, although the actual concentrations were reportedly higher in blood.

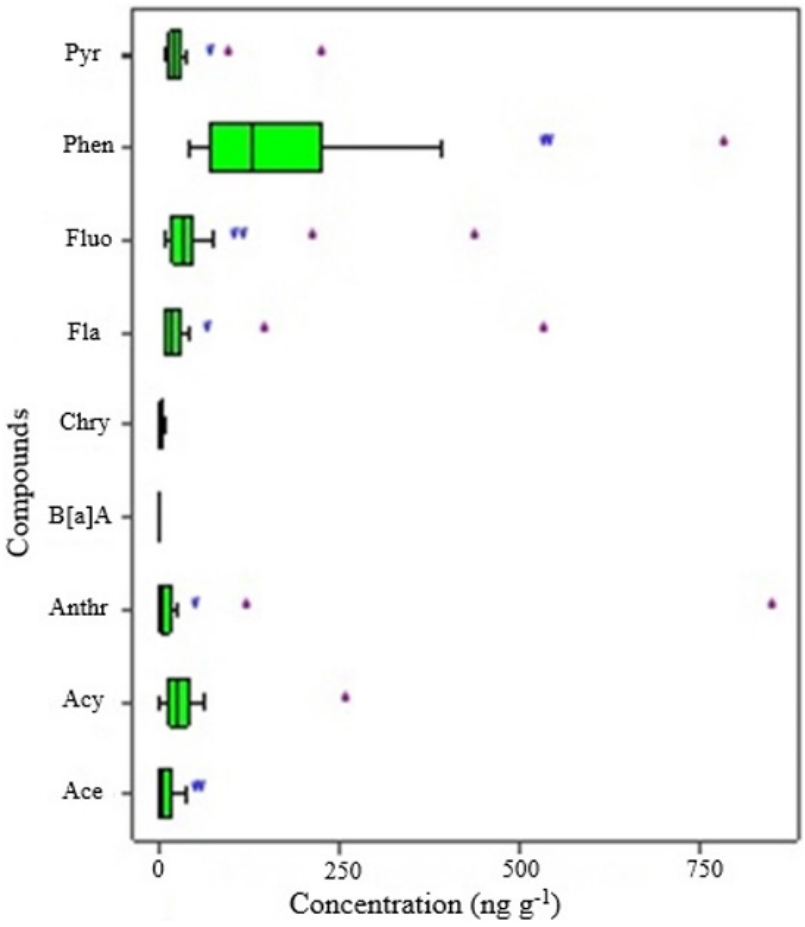

Figure 5. ∑PAH concentrations (ng·g^{-1}) in human hair. PAH: Polycyclic aromatic hydrocarbon.

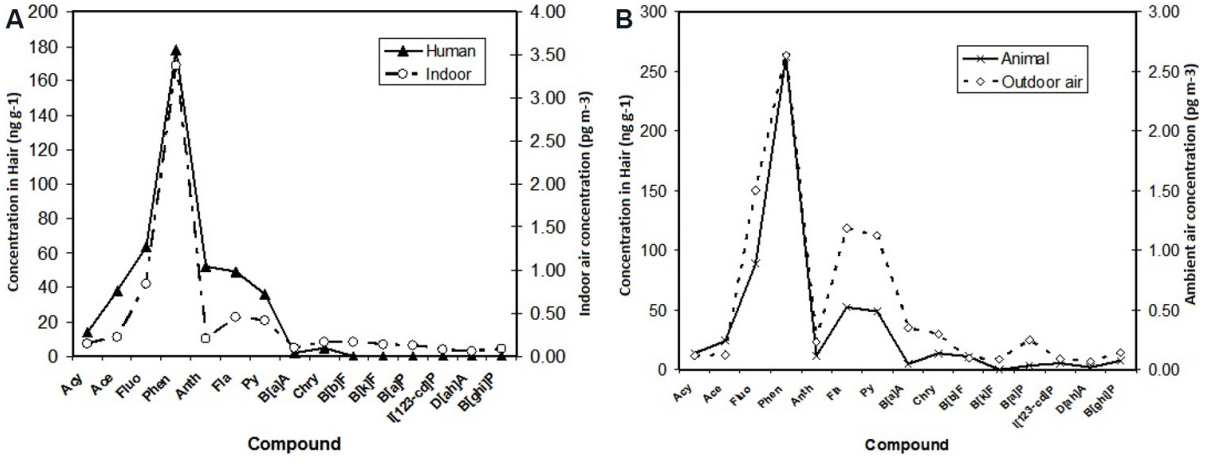

Figure 6. ∑PAH concentration in human hair and indoor air. (B) ∑PAH concentration in animal hair and outdoor air. PAH: Polycyclic aromatic hydrocarbon.

The link between indoor pollution and the concentration of pollutants in hair has received far less attention. Neuber and Merkel[56] investigated the relationship between indoor air concentrations of lindane and DDT from wood preservatives and the concentrations of hair in preschool children in rural Germany. Lindane

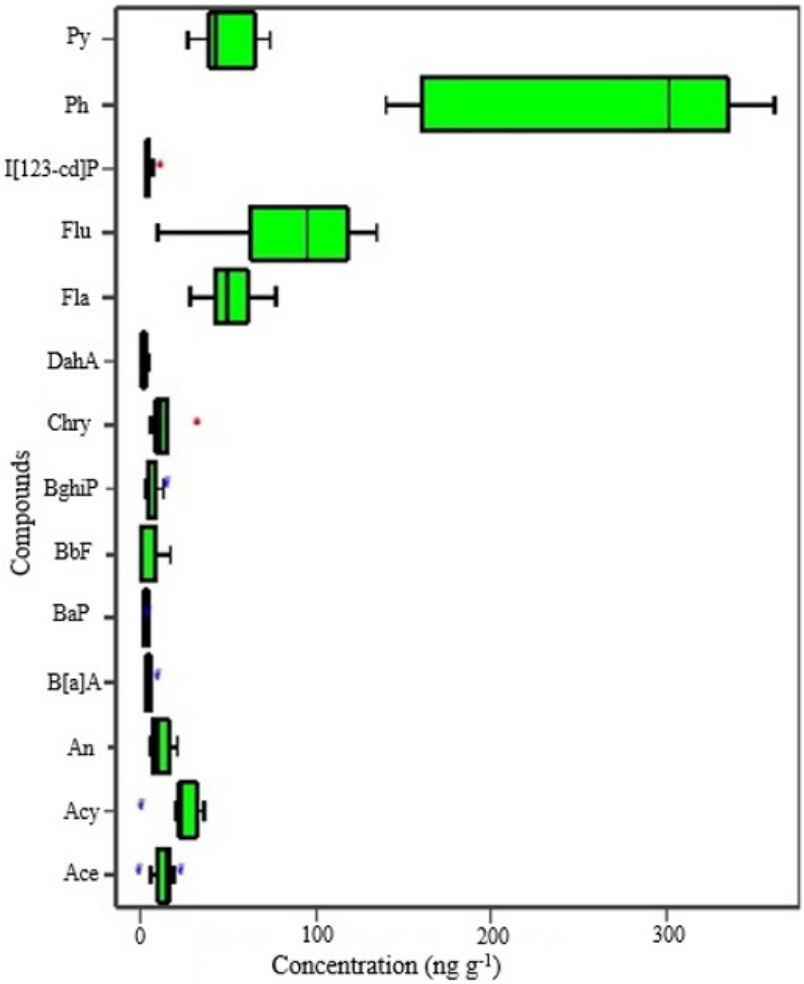

Figure 7. PAH concentrations (ng·g^{-1}) in sheep hair. PAH: Polycyclic aromatic hydrocarbon.

was detected in the vast majority of samples, whereas DDT was present in 30% of the samples, which led them to suggest that hair may be used for assessment of indoor air pollution by lindane and DDT. Kocan *et al.*[41] reported identical isomer patterns for PCDD/Fs in ambient air and hair of individuals scavenging for recyclable waste from a municipal waste dumping site in Cairo, Egypt.

One of the main challenges in the interpretation of hair data is distinguishing between endogenous and exogenous sources of contamination[36,38,40,57]. It has been argued that the current hair washing procedures designed to remove "external contamination" from hair cannot distinguish between the two[37,38]. The question that needs to be answered is whether semi-volatile organic compounds in the hair are primarily exogenous or endogenous in origin. The approach to addressing this issue has been to wash hair using different approaches as no standardized procedure has been agreed upon. Hair washing procedures to distinguish exogenous and endogenous fractions of hair have included washing with hot water[31,36,37,39] or washing with water followed by hair shampoo[38,55,58]. Altshul *et al.*[38] reported a 25%-35% extraction of POPs following a shampoo wash, whereas Nakao *et al.*[55] found that 50% of PCDD and 65% of PCDFs were removed from hair using the same procedure. At the other extreme, Ostrea *et al.*[58] noted that no significant differences were observed between paired hair samples, before and after washing, for propoxur to varying percentage removal for other pesticides. It is almost impossible to distinguish between the two using the

current methods, and any distinction on the basis of washing is merely operational. Hair has been used extensively in forensic analyses, especially in the field of poisoning[59] and in the diagnosis of certain diseases and nutritional assessments of humans[60-62]. However, there are many studies on trace metals in hair where levels were not consistent with the nutritional status and clinical symptoms of the individuals and did not match other biological indicators (e.g., whole blood, serum, and urine), further putting into question the notion that hair may be reflective of body burden[60].

CONCLUSIONS

Most hydrocarbons, including POPs, are subject to metabolic transformation, which converts them to more polar moieties that can be easily excreted. It has been suggested that, since the hair root is vascularized during growth, pollutants in blood (which are yet to be metabolized) may enter the roots and be stored. However, it is not known what fraction of the body burden actually escapes as intact compounds and becomes stored in hair and other body tissues. In this study, very significant correlations were found between concentrations of both PBDEs and PAHs in hair and ambient air. Since humans spend in excess of 90% of their time indoors, especially in hot arid countries such as Kuwait, the concentrations in human hair were correlated with concentrations of these chemicals in indoor air measured using polyurethane foam passive air samples. Pearson correlation coefficients were very significant for PBDEs (P-value < 0.004) and PAHs (P-value < 0.001). In the case of sheep, the concentrations found in hair were correlated with outdoor concentrations measured using the sample PUF air samplers. The correlations were even better, with P-values < 0.001 for both PBDEs and PAHs. The compound/congener compositions were also nearly identical between hair and ambient air. Correlations in this study are very strong relative to those reported between hair and body tissues. It can be argued, therefore, that the information carried by hair is more reflective of ambient concentrations rather than the body burden of parent compounds. Although the scope of this study is limited, it provides powerful preliminary evidence that hair can be used as a medium to determine ambient air concentrations of POPs and to increase spatial coverage of POPs from areas where there have been no studies conducted. Owing to the ease of sampling and the fact that no special skills are required for sample collection, more such studies should be undertaken to better assess if hair can potentially be used in the regional monitoring of POP contamination, especially from areas where there are no data available and no monitoring networks for POPs exist.

DECLARATIONS

Acknowledgments

Authors acknowledge the support of Kuwait Institute of Scientific Research for funding this research. Authors are thankful to Mr. Jamal Zafar and Mr. Khaled Al-Matrouk for their assistance in chemical analyses.

Authors' contributions

Made substantial contributions to conception and design of the study and performed data analysis and interpretation: Gevao B, Uddin S

Performed data acquisition, as well as provided administrative, technical, and material support: Al-Bahloul M, Al-Mutairi A

Availability of data and materials

The data is available in the report and as an additional Report at Kuwait Institute for Scientific Research. Additional data and information can be made available at request from individuals interested.

Financial support and sponsorship

The study was supported by Kuwait Institute for Scientific Research. The funding body has no role in design and execution of this study.

Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Consent was taken from participants.

Consent for publication

Not applicable.

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