Int Arch Occup Environ Health (2001) 74: 91–101

ORIGINAL ARTICLE

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Internal exposure to PAHs of children and adults living in homes with parquet flooring containing high levels of PAHs in the parquet glue

Received: 9 June 2000 / Accepted: 31 October 2000

Abstract PAHs form during the incomplete combustion of organic substances and hence they are distributed ubiquitously in the environment. PAHs in the diet are the main source of exposure in man. In 1997 a new source of potential PAH exposure was discovered: very high levels of polycyclic aromatic hydrocarbons (PAHs) and benzo-a-pyrene (BaP) were detected in household dust from former American Forces housing in Frankfurt am Main, Germany, built in 1955/1956. This contamination was caused by a parquet glue containing coal tar, the use of which was formerly standard building practice in Germany. Because the inhabitants of these flats were very concerned about the effects on their health, they were offered the opportunity to take part in biomonitoring examinations to assess individual internal PAH exposure. ***Participants and methods:*** 1213 inhabitants from 511 flats/houses took part in the investigation; this corresponds to 12.7% of the total inhabitants of the former US housing estates in Frankfurt am Main. Spot urine samples were collected and frozen until analysis for 1-, 2-, 3- and 4-hydroxyphenanthrene and 1-hydroxypyrene, which was carried out using a very sensitive and practical high performance liquid chromatographic method with fluorescence detection, approved by the Deutsche Forschungsgemeinschaft. ***Results:*** The level of internal exposure to PAHs in study participants living in homes where parquet glue containing PAHs had been used did not differ from the levels found in particpants in whose homes PAH-containing parquet glue was not used. This was true for the whole group as well as subgroups divided according to age and smoking behaviour. Internal exposure was not influenced by different levels of external exposure (BaP in parquet glue and in household dust). Spearman rank correlations between the level of BaP in parquet glue and in household dust were low and insignificant. ***Conclusion:*** An increase in internal PAH exposure due to high levels of PAHs in parquet glue and household dust could not be detected. This confirms earlier results with small collectives, where only low and insignificant trends were found towards higher internal PAH exposure caused by contaminated homes. Therefore, the ad-hoc working group of the Commission for Indoor Air Quality of the German Federal Environmental Agency stated that a definitive threshold limit value cannot be defined for PAH contamination in parquet glue and household dust. For reasons of disease prevention, however, BaP contamination in household dust exceeding 10 mg/kg should be minimised

Key words Polycyclic aromatic hydrocarbons (PAHs) · Parquet glue containing PAHs · PAHs in household dust · Internal PAH exposure · Urinary hydroxypyrene · Urinary hydroxyphenanthrenes

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Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of several hundred substances, made up of two to six condensed benzene rings with molecules containing only carbon and hydrogen atoms. In accordance with the EPA convention, normally 16 different compounds are analysed. Eleven of these were classified as carcinogenic in animal experiments by the Deutsche Forschungsgemeinschaft, DFG, Commission for the Investigation of Health Hazards of Chemical Compounds in the Work Area (Deutsche Forschungs Gemeinschaft, 1999).

Benzo(a)pyrene (BaP), one of the most potent carcinogens among the PAHs, was classified by IARC in Group 2A (for substances probably carcinogenic to humans) (IARC, 1983). PAH inhalation may cause lung cancer and dermal exposure may cause skin cancer in man.

PAHs form during the incomplete combustion of wood, oil, coal and gas, and also other organic substances e.g. during the roasting of meat and smoking of tobacco. Thus they are distributed ubiquitously in the environment. They are detected in the air, in the soil, in water, in plants and also in food. Tobacco smoke, which contains up to 150 different PAHs, has been shown to be the predominant source of these compounds in indoor air.

The main source of exposure to PAHs is the diet, with daily PAH ingestion of 1–16 μg and a mean daily BaP ingestion of about 0.1–0.2 μg/d, and even up to 1–2 μg/d in some cases (Buckley et al., 1995; deVos et al., 1990; Dennis et al., 1983; Lioy et al., 1988; Lodovici et al., 1995; Menzie et al., 1992; Santodonato et al., 1981; Vaessen et al., 1988). PAH/BaP exposure via inhalation is comparatively low, except in smokers, whose level of exposure from smoking may be in the same range as exposure via the diet.

In 1997 a new source of potential PAH exposure was discovered: very high levels of polycyclic aromatic hydrocarbons (PAHs) and benzo(a)pyrene (BaP) were detected in household dust from former American Forces housing in Frankfurt am Main, Germany, built in 1955/1956; however, PAH (BaP) levels in suspended particles in the indoor air were not increased. Further investigations revealed that the contamination of the household dust samples was caused by a parquet glue containing coal tar, the use of which was formerly standard building practice in Germany. These glues were taken off the market in about 1972, when other and better adhesives were introduced. The local health department duly informed all other public authorities and ministeries in Germany about the potential health hazard caused by the former use of PAHs in parquet glues in Germany. This problem had, up till then, not been recognised.

In accordance with the importance of the problem, experts in the field met at two meetings arranged by the Umweltbundesamt, Germany, and special recommendations were published (Federal environmental agency, 1998a, b). In this scheme, the first task was to investigate the parquet glue. If the glue was found to be "dark" it may be a bitumen or coal tar-based glue, and should be analysed for PAHs (BaP). If the level of BaP in the parquet glue was found to be below 10 mg/kg, the material used was bitumen, and no more analyses are necessary. If the level of BaP in the glue was found to exceed 10 mg/kg, a coal tar-based glue was used and household dust samples should be analysed. In cases where the level of BaP in the glue was found to be above 3000 mg/kg, analysis of the household dust samples should be followed by the analysis of indoor air samples. The Federal Environmental Agency recommended a special wiping technique using foam to sample household dust from the surface of the floor. This technique was considered suitable for evaluating the potential exposure to children and excluded old dust or small particles from cracks in the parquet floor being included in the sample; this had been observed in some household dust samples collected by vacuum cleaner. To reduce the potential PAH exposure of the inhabitants, short-term measures were to be carried out if the BaP contamination exceeded 10 mg/kg household dust or 3 ng/m^3 in the indoor air. The experts suggested that, in accordance with national and international practice, BaP should be analysed as an indicator for all PAHs. In accordance with these recommendations, thousands of investigations were carried out in the following months in homes with parquet flooring from the 50s und 60s all over Germany, especially in former US housing areas.

In Frankfurt am Main the former US housing estates comprise about 2800 flats and nearly 100 houses. After the American families moved out, the flats were rented out and the houses were sold, especially to families with small children. In many houses the new owners ordered household dust samples collected by vacuum cleaner to be analysed, but to our knowledge the parquet glue was not tested in most cases. All the flats owned by two big societies were tested for BaP in the parquet glue and household dust, the latter collected using the special wiping method recommended by the Federal Environmental Agency, Germany. As the occupants of the former American Forces housing were very concerned about potential health risks, they were offered the opportunity of receiving consultation arranged by the local health department of Frankfurt am Main and taking part in biomonitoring examinations for PAH metabolites.

The data obtained from ambient monitoring (BaP in household dust) and biological monitoring (metabolites of phenanthrene and pyrene in urine samples) will be reported here.

Participants, material, methods

The possibility of consultation and biomonitoring was first offered during an information evening organised for the inhabitants, following a first meeting of the experts, on February 5th 1998 in Frankfurt am Main. In addition, the inhabitants were informed via the mass media and the distribution of leaflets. All inhabitants of the former US housing estates – flats and houses – were invited to take part in the tests – without any preclusion criteria.

The total number of persons living on the former US housing estates at the time of the investigation was 9548. By December 1998, 1213 persons living in 511 flats/houses had visited the consultation services and given urine samples for analysis for PAH metabolites; this number represents 27% of the children <6 years of age, 22% of children aged 6–12 years, and about 10% of all other age groups – with the exception of young adults (20–<30 years of age) and the age group ≥ 50 years old.

Most of the participants had already been tested before September 1998, when the house owners published all the ambient monitoring results of the flats. Until then, the inhabitants had not been aware of the PAH contamination in the parquet glue used in their homes. Most of them had not been aware of the levels of BaP in household dust samples either, except those persons who had ordered analyses of their own household dust samples.

Biomonitoring

The spot-urine samples were frozen and sent to the Institute of Occupational, Social and Environmental Medicine of the University of Erlangen-Nürnberg/Germany. There they were analysed for 1-, 2-, 3- and 4-hydroxyphenanthrene and 1-hydroxypyrene using a very sensitive and practical high performance liquid chromatographic (HPLC) method with fluorescence detection After an enzymatic hydrolysis of the urine sample PAH-metabolites were selectively adsorbed to a stationary phase carrying Cu-phthalocyastine. The metabolits were then eluted and online transferred to the analytical column (Lintelmann et al., 1994 and 1998). The detection limit of the method is 5 ng metabolite/l urine. The data were calculated on a creatinine basis (ng/g creatinine). The SPSS program, version 8, was used for statistical analyses.

Parquet glue

Three institutes took samples of parquet glue from the flats owned by the two big companies. To do this, a small hole was bored in the parquet flooring, the wood was removed and a sample of the glue was taken. Afterwards the hole was closed again. The glue was analysed by gas chromatography/mass spectrometry; the limit of detection was 1 mg BaP/kg glue. Here we report on data from 384 flats/houses.

Household dust

Dust samples from private households were collected by the house owners; most of them used their own vacuum cleaners for sampling. In the case of the flats owned by the two big companies, three institutes were commissioned to take household dust samples using the special wiping method recommended by the Federal Environmental Agency. A polyurethane foam was wiped across the floor. The dust swept together on the floor and the dust remaining on the foam were combined to form the household dust sample. The samples were analysed for BaP in three laboratories by gas chromatography and mass spectrometry, with a limit of detection of about 0.1 mg/kg household dust. Data is available from 380 flats/houses.

Results

BaP in parquet glue

All the flats owned by the two companies were tested to determine the levels of BaP in the parquet glue. About one third of these samples yielded values below 10 mg BaP/kg glue, indicating the glue contained bitumen material according to the classification of the Federal Environmental Agency; one third were in the range of 10– < 3000 mg/kg and one third above 3000 mg/kg. In 384 of the 511 flats studied, BaP contamination was found in the parquet glue: in 29% of the flats the level of BaP was < 10 mg/kg; in 33% the level of BaP was in the range 10– < 3000 mg/kg, and in 38% it was > 3000 mg/kg.

Thus there is no evidence that the level of BaP in the glue used in the flats reported here differs from the levels found in all the flats in the former US housing areas.

BaP in household dust

All the flats owned by the two companies were tested to determine the levels of BaP in the household dust sampled using the method recommended by the Federal Environmental Agency. In about 60% of these samples the BaP level was below the detection limit, and in about 90% the BaP level was below 1 mg/kg. About 10% of the samples contained more than 1 mg BaP/kg, and in 1.5% the BaP level exceeded the "limit value" of 10 mg/kg. In 380 of the 511 flats studied, BaP contamination was found in the household dust: in these flats the BaP level was in the same range as in all the flats of the two companies. Thus there is no evidence that the BaP levels in the flats reported here differ from the levels found in all the flats in the former US housing areas.

There was little correlation between the levels of BaP in parquet glue and in household dust (not significant).

PAH metabolites in urine

Table 1 gives the results for 1-, 2-, 3- and 4-hydroxyphenanthrene, and 1-hydroxypyrene in all urine samples ($n = 1213$), listed according to the different age groups. PAH metabolite concentrations per g creatinine were significantly higher in the urine specimens from children below the age of 6 years old than in those from all other age groups. On a litre basis this difference decreased, but was still significant. On the other hand, in adults the PAH metabolite concentration in urine did not differ between the age groups, so that adults ≥20 years are grouped together.

Table 2 summarises the data for different age groups in relation to the BaP levels in the parquet glue according to the classification of the Federal Environmental Agency, i.e. < 10 mg/kg, 10– < 3000 mg/kg, and > 3000 mg/kg. In statistical tests (Kruskal-Wallis) the differences between the levels of internal exposure were not significant in the study participants as a whole, nor in the group of children below 6 years including a subgroup of children 0.5–2 years old (data not shown) and the group of adult non-smokers (Fig. 1a, b).

Table 3 shows the biomonitoring results for different age groups in relation to the BaP levels in the household dust. Here too, no significant differences were seen between the three groups including the subgroup of children 0.5–2 years old with different external exposure resulting from the different levels of BaP contamination in household dust. (Fig. 2a, b).

All Spearman rank correlations between the different metabolites were significant ($P < 0.001$). Spearman rank correlations between the levels of BaP in parquet glue and in household dust and PAH metabolites excreted by the inhabitants were low and insignificant (Table 4). Smoking led to significantly increased levels of internal exposure, except for 1-hydroxyphenanthrene (Table 4).

Discussion

In 1997 a "new", previously unknown indoor source of PAHs was detected: coal tar parquet glues, the use of

Table 1 PAH metabolites in 1213 inhabitants of the former American Forces housing estates in Frankfurt am Main, Germany

	n	mean ± SD ng/g crea	range ng/g crea	P 5 ng/g crea	P 25 ng/g crea	P 50 ng/g crea	P 75 ng/g crea	P 95 ng/g crea
1-OHPhen								
0–< 6 y	347	543 ± 375	LOD-2887	176	318	445	653	1210
6–< 12 y	261	404 ± 283	LOD-2843	143	238	341	475	892
12–< 20 y	110	409 ± 258	10–1467	139	249	334	495	962
≥20 y all	495	439 ± 331	LOD-3075	131	252	351	530	1012
≥20 y NS	289	445 ± 358	LOD-3075	119	247	350	521	1103
≥20 y S	131	431 ± 285	LOD-2290	153	263	371	541	830
2-OHPhen								
0–< 6 y	347	293 ± 255	LOD-2588	90	160	218	327	768
6–< 12 y	261	227 ± 164	LOD-1464	75	126	180	272	518
12–< 20 y	110	253 ± 221	56–1745	75	126	174	294	574
≥20 y all	495	274 ± 214	LOD-2395	90	154	223	317	631
≥20 y NS	289	266 ± 231	LOD-2395	82	144	206	306	625
≥20 y S	131	289 ± 197	LOD-1209	92	173	232	355	685
3-OHPhen								
0–< 6 y	347	455 ± 321	LOD-2687	134	262	367	539	1136
6–< 12 y	261	374 ± 210	43–1272	117	233	334	448	796
12–< 20 y	110	343 ± 234	60–1789	108	183	290	430	791
≥20 y all	495	340 ± 237	LOD-1850	105	185	279	424	794
≥20 y NS	289	305 ± 209	LOD-1371	102	173	244	384	680
≥20 y S	131	426 ± 284	LOD-1850	122	228	364	562	927
4-OHPhen								
0–< 6 y	347	95 ± 197	LOD-2613	LOD	23	47	103	310
6–< 12 y	261	51 ± 59	LOD-420	LOD	18	34	58	172
12–< 20 y	110	63 ± 138	LOD-1007	LOD	18	30	55	216
≥20 y all	495	58 ± 148	LOD-2600	LOD	14	33	62	151
≥20 y NS	289	58 ± 170	LOD-2600	LOD	12	30	62	147
≥20 y S	131	61 ± 130	LOD-1327	LOD	19	37	61	177
1-OHPyr								
0–< 6 y	347	187 ± 158	LOD-1429	LOD	97	148	232	467
6–< 12 y	261	142 ± 118	LOD-833	27	69	118	173	331
12–< 20 y	110	127 ± 77	LOD-407	38	74	106	152	298
≥20 y all	495	122 ± 117	LOD-1172	22	56	88	142	362
≥20 y NS	289	100 ± 101	LOD-1172	23	51	77	112	262
≥20 y S	131	173 ± 137	LOD-716	24	78	135	239	472

NS: non smoker; S: smoker; LOD: limit of detection

which had been standard construction practice in Germany in the 1950s and 1960s. In consequence, all flats from the former American Forces housing estates in Frankfurt am Main, Germany, were tested for the BaP levels in the parquet glue and household dust. No correlations could be found between the levels of BaP in the parquet glue and the household dust samples or indoor air contamination. These results did not differ from those obtained in the flats of the participants of our study. Thus, it appears that the results from the flats reported here are representative of all flats, irrespective of the fact that the participants volunteered to be examined.

The results reported here correlate well with the data published by the Federal Environmental Agency, Berlin, Germany (Dieckow et al., 1999); no correlations could be found between the levels of BaP in parquet glue, household dust, and indoor air in more than 5000 sets of data which included both German flats and former US housing from all over Germany. In Baden-Württemberg a specific study was conducted by the Public Health Department (Public Health Department, 1999): increased levels of BaP in indoor air and household dust samples from homes where parquet glue containing BaP had been used were rare, and no correlations could be detected between the levels of BaP in the different media. For other, more volatile polycyclic aromatic hydrocarbons such as phenanthrene, however, correlations between the BaP concentration in parquet glue and volatile PAHs in the other media were confirmed. The most important factor governing potential indoor contamination with PAHs from parquet glue containing PAHs therefore seems to be the condition of the parquet flooring and its basis.

Especially infants and small children were considered to be at risk from exposure to PAHs in the parquet glue due to the ingestion and/or dermal absorption of contaminated household dust while crawling and playing on the floor. According to several studies, infants and children ingest a mean daily amount of earth or household dust of about 20 mg with a p95 value of 100 mg/day (Calabrese et al., 1989; Finley et al., 1994). Assessment of the exposure–calculated using the mean and maximum values for the household dust analysed in 1997 (see introduction)–showed that given daily inges-

Table 2 PAH metabolites in urine of inhabitants of the former US housing estates – in relation to the levels of BaP in the parquet glue used in their flat

	< 10 mg BaP/kg glue			10– < 3000 mg BaP/kg glue			≥3000 mg BaP/kg glue			Kruskal-Wallis test
	n	mean ± SD ng/g crea	med ng/g crea	*n*	mean ± SD ng/g crea	med ng/g crea	*n*	mean ± SD ng/g crea	med ng/g crea	
1-OHPhen										
all	266	433 ± 353	333	302	485 ± 392	397	382	436 ± 272	372	0.110
0– < 6 y	81	538 ± 411	427	87	537 ± 430	418	91	536 ± 354	467	0.737
6– < 12 y	54	391 ± 410	300	70	401 ± 301	316	93	431 ± 214	370	0.016
12– < 20 y	24	411 ± 254	362	24	474 ± 351	374	48	349 ± 185	304	0.360
≥20 y all	107	386 ± 278	304	122	499 ± 417	404	150	405 ± 255	350	0.019
≥20 y NS	63	389 ± 257	307	71	508 ± 458	408	88	432 ± 289	364	0.261
≥20 y S	35	360 ± 295	295	29	485 ± 415	397	33	438 ± 196	420	0.018
2-OHPhen										
all	266	265 ± 231	215	302	286 ± 246	217	382	252 ± 188	198	0.416
0– < 6 y	81	322 ± 348	227	87	307 ± 247	220	91	279 ± 210	222	0.856
6– < 12 y	54	307 ± 159	203	70	229 ± 176	162	93	236 ± 183	185	0.593
12– < 20 y	24	216 ± 126	174	24	311 ± 254	216	48	231 ± 251	156	0.223
≥20 y	107	250 ± 154	219	122	299 ± 277	234	150	253 ± 151	207	0.449
≥20 y NS	63	235 ± 147	203	71	325 ± 329	225	88	241 ± 130	197	0.280
≥20 y S	35	253 ± 161	231	29	269 ± 222	230	33	308 ± 202	232	0.563
3-OHPhen										
all	266	350 ± 226	296	302	404 ± 314	333	382	374 ± 245	320	0.153
0– < 6 y	81	439 ± 286	362	87	475 ± 413	363	91	439 ± 261	368	0.964
6– < 12 y	54	353 ± 247	305	70	365 ± 206	339	93	415 ± 216	366	0.048
12– < 20 y	24	281 ± 135	271	24	468 ± 356	353	48	306 ± 183	257	0.037
≥20 y	107	299 ± 151	280	122	364 ± 269	295	150	331 ± 256	251	0.275
≥20 y NS	63	280 ± 153	257	71	343 ± 260	283	88	295 ± 198	237	0.544
≥20 y S	35	336 ± 142	325	29	441 ± 345	371	33	469 ± 359	373	0.409
4-OHPhen										
all	266	65 ± 139	34	302	74 ± 136	41	382	61 ± 151	34	0.163
0– < 6 y	81	88 ± 200	42	87	106 ± 168	54	91	99 ± 276	48	0.504
6– < 12 y	54	52 ± 57	32	70	53 ± 56	41	93	54 ± 60	34	0.747
12– < 20 y	24	104 ± 209	39	24	51 ± 50	41	48	51 ± 140	25	0.063
≥20 y	107	48 ± 80	27	122	67 ± 152	36	150	46 ± 50	32	0.335
≥20 y NS	63	43 ± 67	27	71	68 ± 125	33	88	49 ± 57	33	0.287
≥20 y S	35	58 ± 106	31	29	80 ± 243	30	33	47 ± 31	37	0.250
1-OHPyr										
all	266	159 ± 150	112	302	151 ± 113	128	382	143 ± 131	115	0.298
0– < 6 y	81	211 ± 147	184	87	181 ± 124	150	91	192 ± 196	152	0.244
6– < 12 y	54	171 ± 192	91	70	124 ± 78	112	93	150 ± 83	145	0.069
12– < 20 y	24	106 ± 70	97	24	157 ± 76	152	48	121 ± 79	101	0.007
≥20 y	107	125 ± 131	92	122	144 ± 124	102	150	115 ± 109	85	0.093
≥20 y NS	63	109 ± 146	80	71	133 ± 120	79	88	88 ± 59	78	0.118
≥20 y S	35	141 ± 97	126	29	194 ± 144	156	33	192 ± 155	134	0.326

NS: non-smoker; S: smoker

tion of 20 or 100 mg household dust and complete absorption, children may ingest up to 10–140 μg PAH per day, or up to 0.50–9 μg BaP, respectively. This would exceed several fold the "normal" PAH exposure in adults resulting from dietary intake, which has been investigated in different studies in several countries (Buckley et al., 1995; deVos et al., 1990; Lioy et al., 1988; Lodovici et al., 1995; Menzie et al., 1992; Santodonato et al., 1981; Vaessen et al., 1988).

The first household dust samples, however, were collected by vacuum cleaner, and in some of them small black particles were found, presumably particles of the old glue material or old dust from inside crevices in the parquet flooring, which was assumed not to be relevant for the children's exposure. When the calculated level of exposure was reassessed using the data from household dust samples gained by wiping the surface of the floor, 100-fold lower values were obtained for the amount ingested by children playing on the floor. Moreover, recent analyses showed that the absorption of PAHs (BaP) from contaminated earth is about 1%, and about 10% when milk powder is given at the same time (Hack et al., 1996). Taking this into account, the probable increase in exposure for children would be only slight. Because of the uncertainties in the calculation methods, however, and the need for individual data for risk assessment and risk communication, internal exposure was analysed in many inhabitants on a voluntary basis.

Ten years ago Jongeneelen proposed 1-hydroxypyrene as an appropriate indicator of PAH exposure and published an analytical method for 1-hydroxypyrene in urine (Jongeneelen 1988). Meanwhile, all over the world,

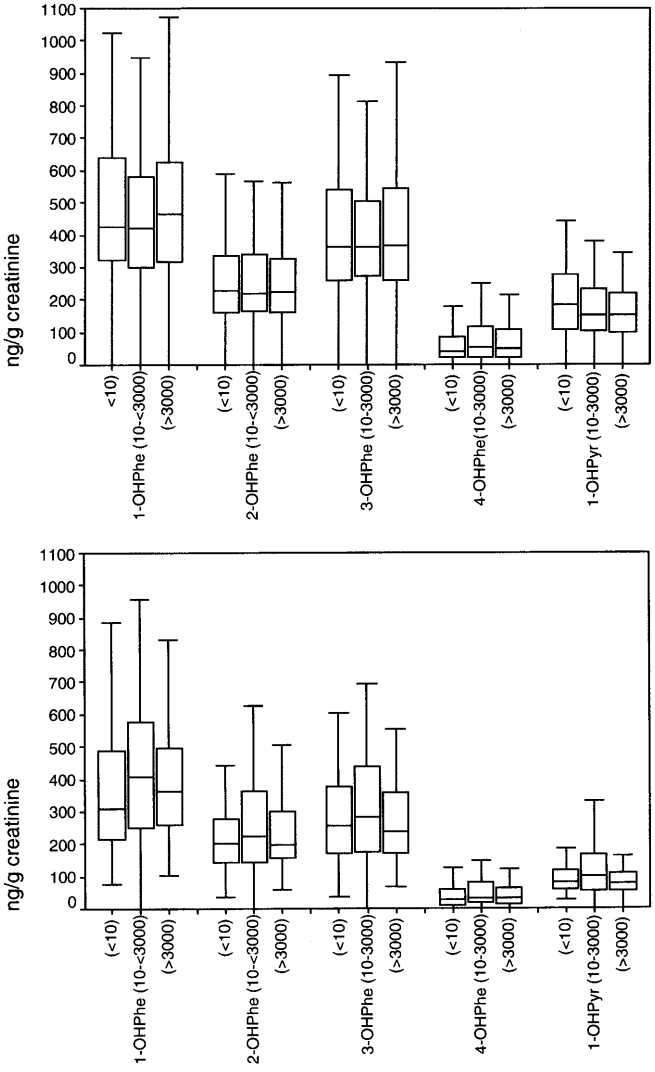

Fig. 1 PAH-metabolites in urine of children (a) and adult nonsmokers (b) in relation to the levels of BaP in the parquet glue used in their flat (mg BaP/kg)

studies of occupational and environmental PAH exposure have been carried out using 1-hydroxypyrene as an indicator of exposure. A lot of studies with 1-hydroxypyrene as a marker of exposure to PAHs have been published, demonstrating not only occupational PAH exposure (Angerer et al., 1997 a, b; Bentsen-Farmen et al, 1999; Feunekes et al., 1997; Jongeneelen., 1998; Jongeneelen et al., 1990; Moen et al., 1997; Nielsen et al., 1996; Ovrebro et al., 1994; Pyy et al., 1997; Santella et al., 1993; Strickland et al., 1996; van Schooten et al., 1995), but also environmental exposure (Gilbert et al., 1997; Gündel et al., 1996; Kanoh et al., 1993). Moreover, this metabolite is found in low levels in most human urine, even in persons without apparent environmental, occupational, or smoking exposure (Angerer et al., 1992 and 1997b; Gilbert et al., 1997; Göen et al., 1995; Grimmer et al., 1993; Gündel et al., 1994; Jongeneelen et al., 1985; Levin, 1995; Nielsen et al., 1996; Omland et al., 1994; Ovrebro et al., 1994; van Schooten et al., 1995). In these studies the validity of 1-hydroxypyrene was confirmed (Jongeneelen et al., 1997; Levin 1995; Strickland et al., 1996).

The disadvantage of the procedure for analyzing 1-hydroxypyrene in urine developed by Jongeneelen (1988) is that it is based on only one substance, a metabolite of pyrene. Pyrene itself is considered not to be carcinogenic, and therefore it would be desirable to analyse specific metabolite(s) of the carcinogenic BaP, for example. In our study we used a HPLC procedure, which allows simultaneous determination of different monohydroxylated phenanthrenes and 1-hydroxypyrene in urine (Lintelmann et al., 1994 and 1998). The detection limit of the individual metabolites is about 5 ng/l. This method has already been used successfully in a study with occupationally exposed workers (Mannschreck, 1997). Moreover, in an recent study investigating environmental PAH exposure in women from an industrial area in Germany, it was concluded that this method is suitable for detecting very sensitively and reliably internal PAH exposure in persons occupationally exposed and in the general population (Gündel et al., 1996).

Before discussing the results, however, some limitations of our study have to be mentioned.

Firstly, these are not the results of an epidemiological study with a representative collective including matched controls. The investigation of an appropriate control group was not possible for financial reasons. Instead, all those people living in the flats who wished to take part in biomonitoring were tested. An internal "not exposed or control group" was formed from people living in flats where parquet glue without increased BaP levels had been used. The disadvantage of this procedure is that classification was carried out after urine sampling ("ex post"). On the other hand, the advantage is that all the investigations were double blind (actually triple blind): neither the probands, nor the physician nor the laboratory staff were aware of the exposure status of the persons tested. Also, with respect to potential PAH exposure, a "self selection bias" can be ruled out for all persons tested before information on the BaP content of the parquet glue was available. Moreover, when the BaP contamination in parquet glue and household dust specimens from the American housing estates in general are compared with the data obtained in the participants homes, it can be said that the values are "representative".

Secondly, the investigation was carried out only after the inhabitants had been informed about the possible PAH exposure due to parquet glue containing PAHs and after the public health department had recommended their cleaning routine be intensified in order to reduce potential PAH exposure for children playing on the floor. Thus, it cannot be totally ruled out that under normal "undisturbed" conditions especially children playing on the floor who ingested contaminated household dust might have had higher exposures before this information was made available. As the information about carcinogenic glue under parquet flooring in homes has been spread all over Germany by the mass media,

Table 3 PAH metabolites in inhabitants related to the levels of BaP in the household dust in their flat

	< LOD mg/kg dust			LOD– < 1 mg/kg dust			≥1 mg/kg dust			Kruskal-Wallis test
	n	mean ± SD ng/g crea	med. ng/g crea	*n*	mean ± SD ng/g crea	med ng/g crea	*n*	mean ± SD ng/g crea	med. ng/g crea	
1-OHPhen										
All	565	450 ± 339	364	263	445 ± 308	372	105	482 ± 384	401	0.596
0– < 6 y	167	544 ± 413	445	64	532 ± 389	423	26	497 ± 307	427	0.814
6– < 12 y	125	433 ± 353	337	60	377 ± 191	333	24	429 ± 240	381	0.758
12– < 20 y	58	402 ± 251	337	26	355 ± 201	263	11	472 ± 378	318	0.683
≥20 y all	215	402 ± 270	322	113	450 ± 315	372	44	504 ± 485	383	0.135
≥20 y NS	130	426 ± 307	344	64	451 ± 279	408	23	578 ± 633	384	0.356
≥20 y S	53	373 ± 170	323	29	473 ± 415	331	13	414 ± 223	384	0.704
2-OHPhen										
All	565	272 ± 231	210	263	260 ± 182	220	105	272 ± 261	201	0.954
0– < 6 y	167	316 ± 308	219	64	274 ± 194	224	26	286 ± 164	221	0.911
6– < 12 y	125	249 ± 190	203	60	216 ± 155	177	24	238 ± 160	170	0.490
12– < 20 y	58	242 ± 240	172	26	270 ± 217	174	11	227 ± 222	156	0.541
≥ 20	215	260 ± 170	212	113	272 ± 178	227	44	294 ± 351	248	0.699
≥20 y NS	130	255 ± 172	200	64	265 ± 169	223	23	361 ± 468	252	0.505
≥20 y S	53	270 ± 178	231	29	309 ± 132	237	13	210 ± 128	213	0.473
3-OHPhen										
All	565	375 ± 277	308	263	380 ± 239	329	105	370 ± 254	315	0.448
0– < 6 y	167	464 ± 363	360	64	425 ± 253	365	26	393 ± 163	374	0.991
6– < 12 y	125	394 ± 239	334	60	370 ± 192	356	24	382 ± 205	333	0.996
12– < 20 y	58	309 ± 174	270	26	364 ± 207	298	11	381 ± 474	288	0.423
≥20	215	314 ± 219	254	113	363 ± 260	296	44	344 ± 254	276	0.224
≥20 y NS	130	292 ± 190	238	64	313 ± 208	271	23	382 ± 307	288	0.547
≥20 y S	53	385 ± 274	345	29	498 ± 363	419	13	302 ± 171	277	0.141
4-OHPhen										
All	565	64 ± 116	35	263	70 ± 187	41	105	76 ± 156	38	0.429
0– < 6 y	167	90 ± 159	46	64	114 ± 326	53	26	120 ± 245	32	0.241
6– < 12 y	125	56 ± 61	34	60	48 ± 47	35	24	58 ± 72	43	0.865
12– < 20 y	58	75 ± 183	29	26	43 ± 42	31	11	50 ± 67	28	0.997
≥20	215	45 ± 62	29	113	63 ± 135	34	44	66 ± 137	42	0.115
≥20 y NS	130	45 ± 63	27	64	58 ± 77	34	23	97 ± 184	67	0.019*
≥20 y S	53	51 ± 71	34	29	85 ± 241	39	13	19 ± 15	21	0.033*
1-OHPyr										
All	565	150 ± 141	109	263	146 ± 113	123	105	156 ± 126	124	0.605
0– < 6 y	167	199 ± 173	157	64	177 ± 127	158	26	204 ± 140	163	0.870
6– < 12 y	125	151 ± 138	118	60	141 ± 97	122	24	141 ± 78	148	0.767
12– < 20 y	58	108 ± 60	102	26	142 ± 83	147	11	158 ± 117	106	0.103
≥20	215	122 ± 20	91	113	131 ± 117	95	44	136 ± 138	102	0.827
≥20 y NS	130	105 ± 118	83	64	110 ± 109	78	23	114 ± 89	93	0.860
≥20 y S	53	167 ± 120	138	29	185 ± 133	142	13	160 ± 186	84	0.383

Spearman correlations; *$P < 0.05$, NS: non-smoker; S: smoker

newspapers, magazines, radio and television, nowhere in Germany will an ''undisturbed'' population now be found. Therefore, the above hypothesis will never be confirmed or ruled out. In view of the generally low-level contamination with BaP of housedust samples gained from the surface of the floor using the modified wiping method and the recalculation of the exposure levels with more realistic assumptions, however, big differences in internal exposure could no longer be expected, irrespective of the cleaning activity of the inhabitants.

The results reported here may be readily compared with the results of other studies from different countries using 1-hydroxypyrene as a parameter of PAH exposure (Table 5). In adult non-smokers and smokers, the 1-hydroxypyrene levels were within the levels found in control groups of other occupational or environmental studies (Angerer et al., 1992 and 1997a, b; Göen et al., 1995; Grimmer et al., 1993; Gündel et al., 1994; Levin et al., 1995; Nielsen et al., 1996; Omland et al., 1994; Ovrebro et al., 1994; van Rooij et al., 1994; van Schooten et al., 1995). In our study the median 1-hydroxypyrene levels in urine specimens from smokers were about 50% higher than in those of non-smokers. This is well in accordance with many other studies (Angerer et al., 1992 and 1997; Gilbert et al., 1997; Göen et al., 1995; Gündel et al., 1994; Herikstad et al., 1993; Hong et al., 1999; Jacob et al., 1999; Jongeneelen et al., 1990; Levin., 1995; Santella et al., 1993; Sherson et al., 1992; Sithisarankul et al., 1997; van Rooij et al., 1994; van Schooten et al., 1995), although in some publications no such effect could be found (Jongeneelen et al., 1988; Martin et al., 1989; Ny et al., 1993; Omland et al., 1994; Zhao et al., 1992).

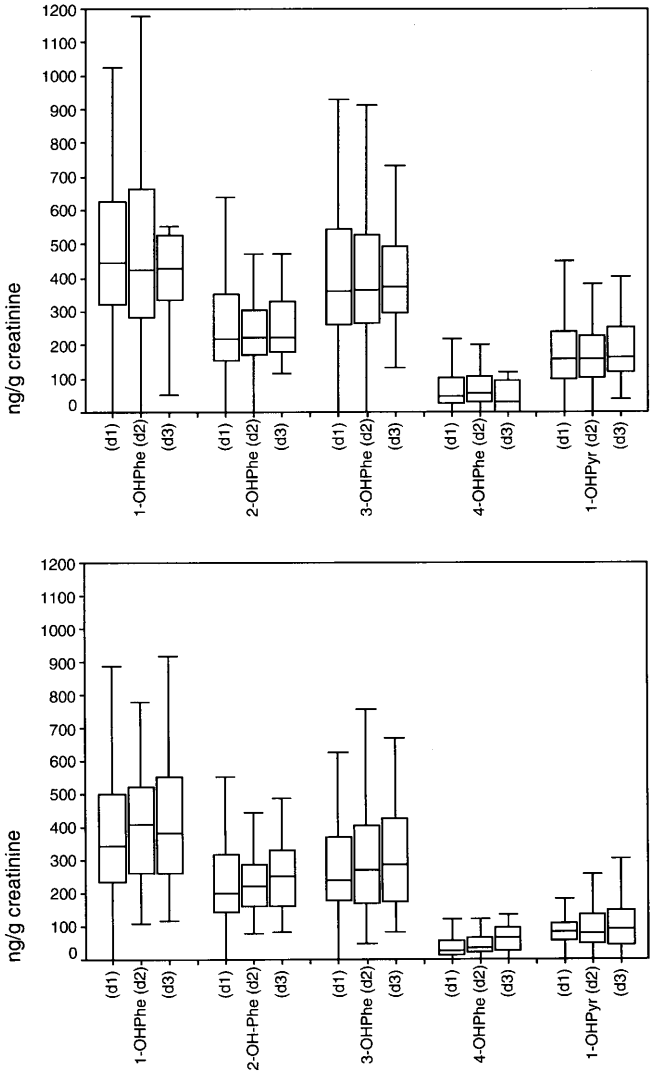

Fig. 2 PAH-metabolites in urines of children (a) and adult non smokers (b) in relation to the levels of BaP in the household dust of their flat (d1: < LOD; d2:LOD– < 1 mg BaP/kg; d3: > = 1 mg BaP/kg)

Significant correlations between internal PAH exposure and the numbers of cigarettes smoked per day were found in our study (Table 4).

With regard to children, to date the 1-hydroxypyrene levels found in 3 groups with a total of 74 children have been published; the 1-hydroxypyrene levels in the urine of the children reported here were well within the expected range, irrespective of the BaP-concentrations found in the parquet glue used in their homes (Table 6). In the international literature, several studies on the internal PAH exposure of children have been published. In the Netherlands, van Wijnen et al., studied 644 children aged 1–6 years old, living in five areas with different levels of PAHs in the soil and ambient air (van Wijnen et al., 1996). The mean and maximum 1-hydroxypyrene levels were found to be 680 ± 980 ng/g creatinine, and 14,000 ng/g creatinine. Similarly high levels of 1-hydroxypyrene were found in the urine of

Table 4 Correlation between PAH metabolites in urine and levels of BaP in parquet glue or household dust and smoking behaviour (cigarettes/day)

Parameter	BaP in parquet gue		BaP in household dust		Cigarettes smoked per day	
	n	*r*	*n*	*r*	*n*	*r*
1-OHPhen	957	0.060	937	0.017	389	0.057
2-OHPhen	957	−0.008	937	−0.002	389	0.168**
3-OHPhen	957	0.041	937	0.021	389	0.344**
4-OHPhen	957	0.031	937	0.016	389	0.118*
1-OHPyr	957	−0.040	937	0.032	389	0.407**

Spearman correlations, two-tailed; $*P < 0.05$; $**P < 0.01$

thirty 8-year-old children living in an industrial town in Poland: the median levels were 300–600 ng/g creatinine, with maximum levels up to 7000 ng/g creatinine (Siwinska et al., 1998). Thus, these children are subject to a much higher level of internal PAH exposure than the children studied from the former American Forces estates in Frankfurt, Germany. It may be assumed, however, that the analytical methods used may differ. In another study with children of 2–4 years of age from low-income families in the USA, mean 1-hydroxypyrene metabolite concentrations of 98 ± 82 ng/g creatinine were found, with a range of 16–360 ng/g creatinine (Chuang et al., 1999). These data correlate well with the exposure data found in our study.

Here, the results of a smaller study with children <6 years of age conducted in Frankfurt am Main, Germany, in December 1997 were confirmed: 60 children living on the former US housing estates were found to have a small but insignificant increase in the level of internal exposure compared to 23 control children living in the same region but not on the former US housing estates (Heudorf et al., 1998 and 1999). Similar biomonitoring results, for 1-hydroxypyrene only, were obtained in another study of children from former US housing areas in Bavaria, Germany. The mean and 95th percentile for the levels of 1-hydroxypyrene excreted in the urine of 50 children with parquet flooring in their homes were 200 and 470 ng/g creatinine, compared to 240 and 450 ng/g creatinine in 29 controls (Lederer et al., 1998). Therefore, the calculated exposure–with daily ingestion of 20 and 100 mg household dust, sampled using vacuum cleaners, and 100% absorption–was not confirmed. The data correspond to exposure calculated using a 10% absorption rate and household dust samples obtained by wiping. To date no other studies have been published with adults from homes with parquet glue containing BaP.

As for the phenanthrene metabolites, to date no other studies conducted with children have been published; our data on adults can be compared with one environmental study with 97 non-smoking and 27 smoking women in Germany from 1994; the urine samples were analysed using the same method and in the same laboratory (Gündel et al., 1996). Relative to this collective, our adult study participants from homes with and without parquet glue containing BaP were found to

Table 5 1-Hydroxypyrene in urine of adults - data from various control groups - in comparison with the data from inhabitants of the former American Forces housing estates in Frankfurt am Main, Germany (ng/g creatinine)

Non-smoker				Smoker				Country	Author, year
n	Mean ± SD	Median	range	*n*	Mean ± SD	Median	range		
10		100	< 50–230	10		400	60–2300	Germany	Angerer et al., 1992
28		120	30–400	21		230	30–1310	Germany	Gündel et al., 1994
14		100	30–240	8		180	60–1300		
49		< 40	< 40–540	20		280	< 40–600	Germany	Göen et al., 1995
14		70		10		170		Sweden	Levin, 1995
13	160							Norway	Ovrebro et al., 1994
97	190	150	60–1560	27	570	480	180–1500	Germany	Angerer et al., 1997c
				16		290		The Netherlands	Jongeneelen et al., 1985
13	92		24–232	7	250		102–564	Canada	Gilbert et al., 1997
26		LOD	LOD-20	42		LOD	LOD-22		Omland et al., 1994
		240	80–580			500	400–1580		Van Rooij et al., 1994
14	700 ± 640			16	940 ± 740			The Netherlands	Van Schooten et al., 1995
12	108 ± 62	100	60–220					Denmark	Nielsen et al., 1996
289	100 ± 101	77	LOD-1172	131	173 ± 137	135	LOD-716	Germany	This study

Table 6 1-Hydroxypyrene in urine from children–data from various control groups, in comparison with data from children living on the former American Forces housing estates in Frankfurt am Main, Germany (ng/g creatinine)

n	mean ± SD	median	P 95	range	Children's age	Country	Author, year
23	168 ± 88	149	329	LOD–334	0– < 6 y	Germany	Heudorf et al., 1998
29	221 ± 140	200	470		0– < 12 y	Germany	Lederer et al., 1998
22		162	309	LOD–425	0– < 6 y	Germany	Angerer, 1999
347	187 ± 158	148	467	LOD–1429	0– < 6	Germany	This study
261	142 ± 118	118	331	LOD–833	6– < 12 y	Germany	This study

have lower internal PAH exposure, as expressed by hydroxylated phenanthrenes and 1-hydroxypyrene; this was true for both smokers and non-smokers. This might be due to the fact that Gündel et al., investigated women living in an heavily industrialised area in Germany, whereas the participants in our study lived in housing areas without heavy pollution in the neighbourhood, neither from industry nor from traffic emissions.

On the basis of this data – ambient and biological monitoring data – the ad-hoc working group of the Commission for Indoor Air Quality of the Federal Environmental Agency, Germany, declared that a definitive threshold limit value, below which there is no risk to the health from exposure to PAH contamination due to coal-tar parquet glues, cannot defined. For reasons of hygiene and disease prevention, however, BaP contamination in household dust above 10 mg/kg should be minimised. In special cases, consultation with an expert in environmental medicine is recommended; this should include the carrying out of biomonitoring analyses. This will be part of the official recommendations for the treatment of homes in which parquet glue containing PAHs was used, which are being prepared by a special working goup from the ministeries for building in Germany and which will be published in the forthcoming months.

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