INDOOR MONITORING OF HOMES WITH WOOL CARPETS TREATED WITH PERMETHRIN

E Berger-Preiß[1*], K Levsen[1], G Leng[2,3], H Idel[2] and U Ranft[4]

[1]Fraunhofer Institute of Toxicology and Aerosol Research, Drug Research and Clinical Inhalation, Nikolai-Fuchs-Str. 1, D-30625 Hannover; Germany
[2]Institute of Hygiene at the University Düsseldorf, D-40225 Düsseldorf, Germany
[3]Bayer AG Leverkusen, Medical Department, D-51368 Leverkusen, Germany
[4]Medical Institute of Environmental Hygiene, University of Düsseldorf, D-40225 Düsseldorf, Germany

ABSTRACT
Textile floor coverings with wool are usually treated with the moth repellent permethrin, an insecticide belonging to the pyrethroid group, to protect against insect damage.To clarify whether this may cause adverse health effects, we investigated the indoor exposure to permethrin in 80 private homes with wool floor coverings (monitoring of permethrin concentrations in wool fibers, house dust, and on suspended particles in the air). Furthermore, three permethrin metabolites were determined in the urine of the inhabitants.The permethrin concentrations found in house dust ranged from <1 to 659 mg/kg. The concentrations observed in the air varied between <1 and 15.2 ng/m³. Correlations between house dust permethrin concentrations and permethrin concentrations in the air were not found. The metabolite concentrations determined in the urine of the inhabitants were in the same range as those of the general population.

INDEX TERMS
Wool floor coverings, permethrin, indoor/biological monitoring, house dust, indoor air concentrations.

INTRODUCTION
Textile floor coverings with wool, woven wool carpets and knotted wool rugs are usually treated with the insecticide permethrin (3-phenoxybenzyl(1-RS)-cis,trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane-1-carboxylate) to protect against damage by moths and creatine-digesting beetle larvae. The recommended application is in the range of 60 – 180 mg permethrin /kg wool (Klingenberger, 1994).

The insecticide permethrin belongs to the group of synthetic pyrethroids. These compounds are derived from naturally occurring pyrethrins, which can be extracted from several chrysanthemum species. In contrast to natural agents, the synthetic analogues are more stable, especially after indoor use (Berger-Preiss, 1997).

Thus, permethrin provides a permanent protection of wool textile floor coverings even after vacuum and dry cleaning. Permethrin is usually applied in the manufacture process of the wool yarn by different procedures (for instance during the yarn dyeing process and the washing of the wool). In these procedures, the insecticide is incorporated into the fiber. A contamination of the indoor environment in homes with wool carpets by abrasion of

* Contact author email: berger-preiss@ita.fraunhofer.de

permethrin-protected fibers is thus conceivable. In consequence, a permethrin contamination of house dust and indoor air via suspended particles may be expected.

Permethrin is known to have a low mammalian toxicity. Nevertheless, different toxicological effects have been observed in animal studies (Aldrige, 1990). For humans, depending on the concentration and the individual susceptibility, a variety of reversible symptoms have been reported such as headache, dizziness, nausea, irritation of the skin, mucosa, and paraesthesia (He, 1988, 1989). No adverse health effects for inhabitants are expected at low permethrin levels observed after indoor application. At the same time, an increasing number of health complaints after indoor use of pyrethroids and in connection with permethrin-protected wool carpets have been reported by the German Federal Institute of Consumer Health and Veterinary Medicine (between 1990 and 1998, 37 of such suspected cases were related to wool carpets (Hahn, 1998)). As a result of this controversial discussion the present study was initiated.

To clarify whether permethrin in treated wool carpets of private homes has an impact on the inhabitants, the following investigations were performed:

- The permethrin concentrations were determined in wool carpets, house dust (sedimented dust) and on suspended particles in the indoor air. (Because of the low vapor pressure of permethrin (45 μPa at 25 ^{o}C) it is unlikely that detectable amounts of permethrin are transferred from the carpet fibers to the gas phase.)
- Suspended particles were collected resolved according to particle size with an Andersen impactor in order to determine which fraction of particle-bound permethrin is respirable.
- The concentrations of three characteristic permethrin metabolites in the urine of the inhabitants were measured.

METHODS

The indoor contamination caused by permethrin was investigated in 80 homes with wool textile floor coverings (wool rugs, wall-to-wall wool carpets) located in or nearby Hanover, Germany. During a 2-year study, house dust was collected once (by vacuum cleaning) and suspended particles were sampled twice (Pallflex filters, sampling ~ 10 m³ at a rate of 2.6 – 3 m³/h). In addition, suspended particles were collected with a seven-stage Andersen impactor (4-10 μm). The samples and carpet fibers (where possible, a piece of the corresponding woolen textile floor covering was investigated) were analyzed for permethrin. Before analysis, the samples were extracted with ethyl acetate in an ultrasonic bath (suspended particles) or in a Soxhlet apparatus (carpet fibers, house dust). Where necessary, the extracts were cleaned up on silica gel mini-columns. Before extraction, the house dust samples were separated into two fractions (dust fraction I: <2mm, dust fraction II: >2mm).

Samples of the 24-hour urine of the inhabitants (145 participants) were collected twice for a determination of the metabolites (cis-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane-carboxylic acid (cis-DCCA), trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylic acid (trans-DCCA), 3-phenoxybenzoic acid (3-PBA)). After hydrolysis, derivatisation and extraction, the samples were analyzed (Kühn, 1996).

The permethrin and metabolite concentrations were determined using a gas chromatography-mass spectroctrometry system in EI-mode. For quantification the selected ion monitoring was used.

All data were evaluated by statistical analysis. For all calculations the SAS software package version 6.12 under Windows NT and Open VMS was used.

RESULTS

In 44 homes, it was possible to obtain samples of the wall-to-wall carpets for analysis. In five of these samples, no permethrin could be detected, while in the remaining 39 samples permethrin could be determined in the wool fibers. The results are shown in Table 1. Values <1 mg/kg (limit of quantitation, LOQ) were considered as 0.5 LOQ for statistical analysis.

Table 1. Permethrin concentrations in wall-to-wall carpets (mg permethrin/kg wool)

samples	minimum	median	90.percentile	maximum
44	<1	37.37	136.00	244.90

As a result of carpet fiber abrasion, permethrin was found in the house dust of most of the investigated homes. In Table 2 the statistical data (dust fraction I <2mm) are shown for all investigated homes. On average, the permethrin concentration in the dust fraction I (<2mm) was 1.3 times higher than that found in the house dust fraction II (>2mm). 42 of the investigated homes were equipped with wall-to-wall wool carpets, 25 with woven or knotted wool rugs and 13 with both. Tables 3 and 4 show the data for homes with wall-to-wall wool carpets and wool rugs, separately. Values <1 mg/kg (limit of quantitation, LOQ) were considered as 0.5 LOQ.

Table 2. Permethrin concentrations in house dust (mg permethrin/kg dust)

samples	minimum	median	90.percentile	maximum
80	<1.00	9.65	129.10	659.15

Table 3. Permethrin concentrations in house dust (mg permethrin/kg dust) of homes with wall-to-wall wool carpets

samples	minimum	median	90.percentile	maximum
42	<1.00	22.25	181.60	659.15

Table 4. Permethrin concentrations in house dust (mg permethrin/kg dust) of homes with woven or knotted wool rugs

samples	minimum	median	90.percentile	maximum
25	<1.00	1.30	18.80	37.30

The permethrin concentrations in house dust of homes with wool rugs were significantly lower than those found in homes with wall-to-wall wool carpets ($p=0.0001$). In 54% of the homes with wall-to-wall textile floor coverings, permethrin was enriched in the dust when compared to the carpet fibers (by a mean factor of 2.3). Furthermore, a significant correlation between the permethrin concentration in the fibers and that found in the dust was observed ($r_{spearman}=0.86$ and $p=0.0001$).

In each of the 80 homes, the indoor air was sampled twice for suspended particles. Table 5 shows the results (permethrin concentrations) of the first sampling (after elimination of one outlier). The values of the second sampling were in the same range. Values <1.0 ng permethrin/m³ air (limit of detection, LOD) were considered as 0.5 LOD. The permethrin

concentrations were in 16.5% (first sampling) and in 21.5% (second sampling) of the samples below the LOD.

Table 5. Permethrin concentrations in the air (suspended particles, ng/m³)

samples	minimum	median	90.percentile	maximum
	<1.00	1.90	5.80	15.20

A statistical analysis of the data demonstrated that there was no correlation between the permethrin concentration in the air (suspended particles) and that found in the house dust and no correlation between the permethrin concentration in the air and the air particle concentration.

Particle size resolved sampling and analysis of permethrin were difficult because of the very low air concentration of permethrin. Thus, sampling of the suspended particles with an Andersen impactor was restricted to three events. It was expected that, because of an increasing specific surface of the particles with decreasing particle diameter, an increasing loading of the particles with permethrin (in mg/kg) would also be observed. Instead, the opposite effect was found in two instances. If the sampled particle diameter decreased from a range of 5.8 – 10 µm to a range of 0.43 – 2.1 µm, the loading decreased from 84.1 to 24.2 mg/kg and from 216.3 to 75.4 mg/kg, respectively, i.e. by a factor of almost 3. For the third sampling, on the other hand, no clear dependence of the permethrin loading on the particle size was observed.

Finally, the urine of the 145 inhabitants was sampled twice and the permethrin metabolites cis-DCCA, trans-DCCA and 3-PBA determined. The metabolite concentrations were below the detection limit in the majority of cases. The median values (for all metabolites) were also below the LOD (0.2 µg/L). For DCCA, the 95.percentiles varied between 0.20 and 0.5 µg/L urine (cis-DCCA) and 0.6 and 0.7 µg/L (trans-DCCA). The 95.percentile for 3-PBA was determined to be 0.9 µg/L.

DISCUSSION

Textile floor coverings (wall-to-wall wool carpets, wool rugs) are protected with permethrin against moths and creatine-digesting beetle larvae. For a reliable protection, a minimum of 35 mg permethrin/kg wool is required, whereby the maximum recommended application is 180 mg/kg (Klingenberger, 1994). The study results were consistent with these recommendations. In 90% of the investigated wall-to-wall carpets, permethrin concentrations were below the maximum recommended concentration and the median value was 37 mg/kg (arithmetic mean 55 mg/kg). Carpet fibers are released into house dust by abrasion. The permethrin concentrations found in the house dust of 80 homes varied between <1 and 659 mg permethrin/kg dust (90. percentile: 129.1 mg/kg). Furthermore, it is known, that a general background level of permethrin is found in many German houses. For the general population a 95.percentile-value of 37.0 mg permethrin/kg dust (Walker, 1999) was reported. The present study demonstrates that permethrin concentrations in house dust are significantly higher in homes with woolen floor coverings.

Resuspended house dust is transported as fine particles in the air. The concentration of permethrin in the air (suspended particles) of the 80 investigated homes was very low and often below the detection limit. The observed statistical correlations and impactor measurements indicate that the indoor air permethrin level (<1 – 15 ng/m³) may be caused by a small fraction of wool fibers (<10 µm) which are suspended in the air.

Finally, three permethrin metabolites were determined in the urine of the inhabitants (145) of the homes with wool textile floor coverings. The 95.percentile varied (depending on the metabolite) between 0.2 and 0.9 µg/L. During the course of the study, data on the background concentration of the pyrethroid metabolites in the urine of the general population in Germany were published (Butte, 1998, Hardt, 1999) and were found to be similar to the metabolite concentrations measured in the present study.

CONCLUSION AND IMPLICATIONS

Wool floor coverings (wall-to-wall carpets, wool rugs) are usually protected against damage with the insecticide permethrin. High concentrations of permethrin were found in the house dust of homes with wool floor coverings due to fiber abrasion; however, the concentrations in the indoor air (suspended particles) were low. The permethrin metabolite concentrations in the urine of the persons living in these homes were in the same range as those of the general population. Thus, if wool carpets contribute at all to the internal burden by pyrethroids, this contribution must be lower than the general internal burden originating from other sources.

ACKNOWLEDGEMENTS

The study was supported by the BMBF (Federal Ministry of Research and Development, Förderkennzeichen 07INR30 B) and the IVA (Industrieverband Agrar).

REFERENCES

Aldrige WN. 1990. An assessment of the toxicological properties of pyrethroids and their neurotoxicity. Critical Reviews in Toxicology. Vol.21, pp 89-104.

Berger-Preiß E, Preiß A, Levsen K et al. 1997. The behaviour of pyrethroids indoors: A model study. Indoor Air. Vol.7, pp 248-261.

Butte W, Walker G, and Heinzow B. 1998. Referenzwerte der Konzentrationen der Permethrinmetaboliten Cl_2CA [3-(2,2-Dichlorvinyl)-2,2 dimethylcyclopropancarbonsäure] und 3-PBA [3-Phenoxybenzoesäure] im Urin. Umweltmed. Forsch. Prax. Vol.3, pp 21-26.

Hahn, A, Michalak, M, Begemann, K, et al. 1998. Ärztliche Mitteilungen bei Vergiftungen nach §16e Chemikaliengesetz. Germany: bgvv-Pressestelle.

Hardt J, Heudorf U, and Angerer J. 1999. Zur Frage der Belastung der Allgemeinbevölkerung durch Pyrethroide. Umweltmed. Forsch. Prax. Vol.4, pp 54-55.

He F, Sun J, Han K, Wu Y et al. 1988. Effects of pyrethroid insecticides on subjects engaged in packaging pyrethroids. Br. J. Industr. Med. Vol.45, pp 548-551.

He F, Wang S, Liu, L, et al. 1998. Clinical manifestations of acute pyrethroid poisoning. Arch. Toxicol. Vol.63, pp 54-58. .

Klingenberger H.1994. Motten- und Käferschutz in Wollteppichen, Proceedings of the VDI Conference ”Luftverunreinigungen in Innenräumen” VDI-report 1122, pp 387-388. Mannheim.

Kühn KH, Leng G, Bucholski KA., et al. 1996. Determination of pyrethroid metabolites in human urine by capillary gas chromatography-mass spectrometry. Chromatographia. Vol.43, pp 285-292.