THE PERSISTENCE OF PYRETHRUM AND CHLORPYRIFOS-ETHYL IN INDOOR ENVIRONMENT - CASE-REPORTS -

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ABSTRACT
Numerous applications for usage of pyrethrum-containing insecticides for indoor use allow the inhabiting of concerning rooms a few hours after bringing out the biocides. Consensus of opinion is, that pyrethrum is no longer detectable within a few days. Correspondingly assumed, after usage of chlorpyrifos the treated rooms show only a residual burden a few days later.

In practice we observe, that pyrethrum can be present for months, chlorpyrifos even for years in indoor environment. In certain cases, e.g. about one year after the pest control, up to 170 mg/kg pyrethrum and up to 1700 mg/kg chlorpyrifos in house-dust can be found. Samples taken from material surfaces show a contamination exceeding 9.000 µg/m^2 for pyrethrum and 43.000 µg/m^2 for chlorpyrifos.

Especially, after an improperly performed pest control in indoor environments a clearly longer-termed exposition of the inhabitants to pyrethrum and chlorpyrifos (and other biocides) as expected was found.

INDEX TERMS
Persistence of biocides, pyrethrum, house dust, wipe-samples, residences

INTRODUCTION
Numerous formulations for controlling household pests are commonly traded. Many insecticidal substances and their formulations are available without restrictions and are allowed to be bought and used by any person without advice (Dullin, Neukirchen and Liedtke 2001). Especially formulations to be sprayed, fogged or vaporized can lead to substance residues on walls, floors and furniture surfaces (Hoffmann 1986; Stolz, Meierhenrich and Krooss 1994; Winter and Hoffmann 2000).
Private, unskilled and professional pest controllers perform pest controls in this way assuming a rapid decay of the biocides after application.
According to a typical recommendation, six hours after usage of a fogger and following ventilation, the treated room can be inhabited as usual (Instructions for use, 2002). Likely advices as well as advertising and popular press information (Brian 1998) implicit a complete decay of the natural pesticide pyrethrum after a few days.

Regularly we examine indoor environment a longer period of time after performing a pest-control. The affected inhabitants develop an impaired health condition, but between determination of causes and clarification of liability and – partly – calling in of a court, several months may pass by.

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Pyrethrum would be not detectable after this time assuming that it has been decomposed. But in fact, we found short-term pesticides like pyrethrum after many months and long-term pesticides like chlorpyrifos-ethyl (short: chlorpyrifos) even after years.

The measurement of surfacial burdens has proved suitable for determination of remaining substance residues after performing pest controls. The background level of non-contaminated surfaces is below detection limits. Based upon our experience, we propose the following provisional scheme for assessment of surface loads:

- less than 10 µg/m^2 = traces
- 10 - 100 µg/m^2 = low burden
- 100 - 1000 µg/m^2 = considerable burden
- 1000 - 10000 µg/m^2 = high burden
- more than 10000 µg/m^2 = very high burden

METHODS

The house dust samples were taken by a vacuum cleaner (according to VDI 4300-8: Measurement of indoor air pollution – Sampling of house dust). The surface contamination was determined by analysing wipe samples. These were taken by repeated wiping of a defined area of the surface with a clean, solvent-soaked cloth. Sampling and analysis was performed according to common procedures described in science literature (Stolz and Krooss, 1993; DFG; Lammers et al. 2001; VDI 1999).

The samples were eluted with ethyl acetate (research grade). Separation, identification and quantification was done by capillary gas chromatography (Shimadzu type 17A) equipped with a mass selective detector (Shimadzu type QP-5000), using an external standard. The samples were injected into a split/splitless injector with an autosampler (type AOC 17).

RESULTS

First case:

A pest controller took measures against fleas with a pure pyrethrum formulation and a chlorpyrifos formulation (application by means of a backpack sprayer with nozzles). Consequently, the inhabitants showed health impairments, thus an expert´s report was ordered by court. The investigated dwelling were inhabited since the application up to the inspection and were regularly cleaned. Samples were taken in various rooms a year after the pest control. Table 1 shows the biocide concentrations in the house-dust samples.

Table 1. Results of the house-dust samples

Substance	*Garden room [mg/kg]*	*Living-room [mg/kg]*	*Kitchen [mg/kg]*	*Corridor [mg/kg]*	*Cellar [mg/kg]*
Pyrethrum	1,4	7	2,6	6,6	42
Chlorpyrifos	14	8	87	120	1.700

detection limits: pyrethrum: 0,1 mg/kg; chlorpyrifos: 0,5 mg/kg.

In order to examine possibly contaminated object surfaces, wipe samples and material samples were taken from various objects. The results of the wipe samples and the surfacial material samples are shown in tables 2 and 3.

Table 2. Results of the material surface samples, calculated as surfacial loads

Sample taken from	*Pyrethrum [µg/m²]*	*Chlorpyrifos [µg/m²]*
Wallpaper, dining-room	1.400	15.200
Wallpaper, kitchen	2.100	43.500

detection limits: 10 µg/m² for pyrethrum and 10 µg/m² for chlorpyrifos

Table 3. Results of the surface examinations (wipe samples)

Sample taken from	*Pyrethrum [µg/m²]*	*Chlorpyrifos [µg/m²]*
Radiator, kitchen	34	770
Radiator, corridor	40	130
Door between kitchen and corridor	130	4.300
Door, dining-room	27	1.600
Sill betw. corridor and garden room	24	2.700
Sill between bathroom and corridor	9.300	33.300
Door frame, stairs to cellar	140	21.700
Skirting-board, kitchen	470	4.300
Floor, corridor	11	6.100

detection limits: 3 µg/m² for pyrethrum and 3 µg/m² for chlorpyrifos

Second case:

The inhabitants of a house sprayed a chlorpyrifos-containing formulation (a commonly available aerosol can) in the upper floor. Several months later, a pest controller used a pyrethrum based formulation against „dust lice“ (lat. Psocoptera) in the whole house (application by means of a backpack sprayer with nozzles). Consequently, the inhabitants complained about affected health. Though taking cleaning measures repeatedly, no health improvement followed, thus one year after the pesticide usage an examination for residues from the pest control was performed. Table 4 shows the biocide burdens in the house-dust samples.

Table 4. Results of the house-dust samples

Substance	*Living-room (downstairs) [mg/kg]*	*Children´s room 1 (upstairs) [mg/kg]*	*Bedroom (upstairs) [mg/kg]*	*Loft (2nd floor) [mg/kg]*
Pyrethrum	1,0	88	170	17
Chlorpyrifos	n.d.	7	7	2

mg/kg = milligram per kilogram n.d. = not detectable

In this case also wipe samples and surfacial material samples were taken for determination of surface contamination. The results of these samples are shown in the following tables 5 and 6.

Table 5. Results of the material surface samples, calculated as surfacial loads

	Pyrethrum		*Chlorpyrifos*	
Sample taken from	*[mg/kg]*	*[µg/m²]*	*[mg/kg]*	*[µg/m²]*
Latex mattress, children´s room 1	200	85.000	4	1.600
Wallpaper, bedroom	7	1.700	n.d.	n.d.

detect. limits: 150 µg/m² (0,5 mg/kg) for pyrethrum / 150 µg/m² (0,5 mg/kg) for chlorpyrifos
n.d. = not detectable.

Table 6. Results of the wipe samples

Sample taken from	*Pyrethrum [µg/m²]*	*Chlorpyrifos [µg/m²]*
Skirting-board, living-room	66	n.d.
Floor, living-room	1.300	n.d.
Couch, living-room	19	n.d.
Shelf, living-room	11	n.d.
High bed, children´s room 1	820	10
Radiator, children´s room 2	33	n.d.
Duplo blocks (toys), children´s room 2	n.d.	34
Stool, loft	1.100	32
Fortress (toy), loft	590	20

detection limits: 3 µg/m² for pyrethrum and 3 µg/m² for chlorpyrifos
n.d. = not detectable

Laboratory experiment: Determination of the decay of chlorpyrifos under living condition.

A common chlorpyrifos-containing insecticidal spray was applied to square glass plates (20 cm edge length). The loaded plates were stored under „dwelling“ condition (15 to 20 °C, protected from direct sunlight) and examined various times after application. Sampling was performed as wipe samples, the analysis procedure being the same as described under „Methods“ except the solvent. Acetone instead of ethyl acetate was used for soaking the wiping cloths and extraction because of further simultaneous investigations (Lammers et al. 2001).
The plates were examined for a period of more than a year; and even 15 months after treatment we found est. 60% of the initial load (see figure 1).

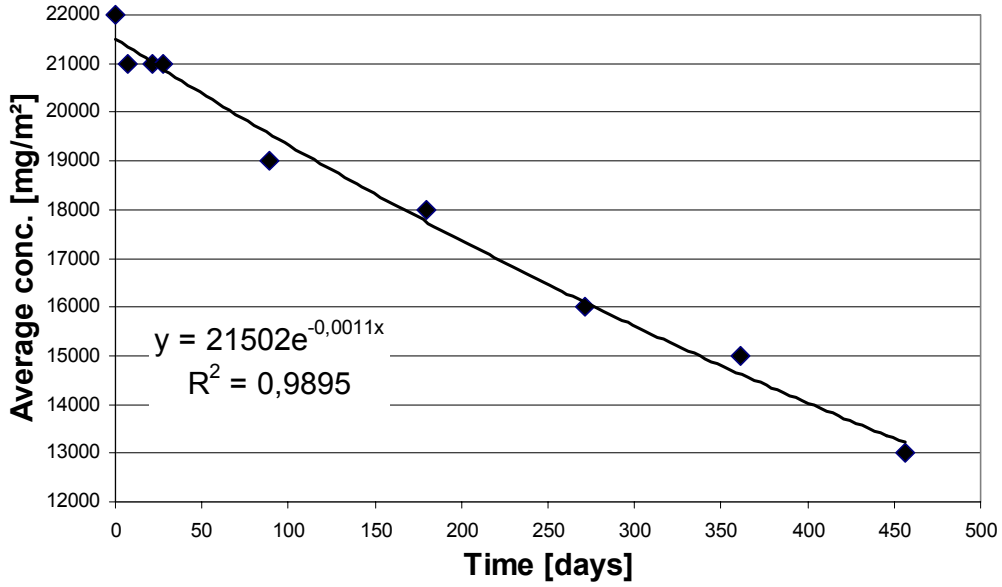

Figure 1. Decay curve for chlorpyrifos, sprayed onto glass plates (Average conc.= 3 samples)

DISCUSSION

In both cases, residues of chlorpyrifos and the short-term biocide pyrethrum were found though samples were taken one year earliest after treatment. The proof is successful both for house-dust and for samples taken from exposed surfaces of various objects. It is impossible to determine the absolute substance amounts applied to the rooms, but obviously darker and colder rooms show higher burdens. Thus it seems likely that the indoor decay rate of the biocides depends both on the usage of the rooms and physical parameters like light and temperature.

Regardless of this we found considerable pyrethrum burdens after 12 – 13 months. These results are in sharp contrast to the common assumption that pyrethrum will completely decompose a few days after indoor usage. The surfacial loads one year after application can range up to some 1000 µg/m² for pyrethrum and even up to some 10000 µg/m² for chlorpyrifos.

Using the assessment scheme described in the introduction, these maximum burdens can be categorised up to high respectively very high. Concentrations in house dust range between 1 mg/kg and 170 mg/kg for pyrethrum resp. 7 mg/kg and 1700 mg/kg for Chlorpyrifos. Concerning pyrethrum, no data for average burdens in house-dust exist. In a study carried out from the German Umweltbundesamt (Friedrich, Becker, Hoffmann et al. 1998), the average house-dust concentration of the commonly used long-term pyrethroid permethrin is 0,22 mg/kg.

During another study, 385 dwellings were investigated. 41 samples contained chlorpyrifos ranging between 0,11 mg/kg and 870 mg/kg (Hostrup, Witte, Hoffmann et al. 1997). The 95-percentile value of house-dust concentrations was determined to 29 mg/kg.

CONCLUSION AND IMPLICATIONS

After user- or professionally performed pest controls, the inhabitants are considerably longer exposed to biocide residues than commonly assumed. The risk of developing health impairments rises with an increasing period of possible substance intake.

Especially, small children take in more biocides orally because they take contaminated toys or other objects in the mouth; besides they absorb more house-dust due to playing behaviour (e.g. creeping on contaminated floor).

Currently, we carry out further investigations concerning the decay of pyrethrum on various surfaces under indoor conditions.

The indoor use of biocides – including pyrethrum – should be newly assessed considering the observed long-term contamination of objects and house-dust.

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