Activities of an Environmental Analysis Van in the German Federal State Schleswig-Holstein

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A cooperation was started between the Union of Physicians of Schleswig-Holstein (Bad Segeberg, Germany) and an environmental engineer in 1992. A mobile unit for environmental analysis was set up, the Environmental Analysis Van (EAV) or mobile umweltambulanz. Inspection of sites and collection of air and dust/material samples for analysis of xenobiotics were performed on request. The results of these cooperation were evaluated to show which sources of indoor pollutants could be particularly relevant to human health impairment. During a 30-month period from July 1993 to December 1995, 1793 site inspections were conducted. Xenobiotic analysis and subsequent advising was performed in 1318 cases; enhanced concentrations of one or more toxic substances (mainly biocides such as pentachlorophenol, permethrin, and/or hexachlorocyclohexane) were found in 71% of the sites analyzed. Formaldehyde, volatile organic compounds, and contamination by molds were also documented. A follow-up was done on 80 clients of the Environmental Analysis Van, which had detected elevated concentrations of permethrin because of pyrethroid-treated carpeting. The effect of removing all contaminated carpeting on health improvement in comparison with nonremoval was statistically significant (p<0.0001). Pyrethroid-treated carpeting, which was already 5, 7, and 10 years old, revealed permethrin concentrations of 115, 100, and 150 mg/kg dust. This result indicates outdoor contamination of permethrin is highly persistent and may be the cause of adverse health effects. Key words: environmental analysis van, environmental medicine, formaldehyde, health effects, hexachlorocyclohexane, indoor air pollution, molds, pentachlorophenol, polychlorinated dibenzodioxins, polychlorinated dibenzofurans, pyrethroids. Environ Health Perspect 105:844–849 (1997) http://ehp.niehs.nih.gov

The value of environmental medicine is controversially discussed in Germany. Although some scientists deny the need for environmental medicine in addition to occupational medicine, an increasing demand for environmental medicine has been observed over the last few years in Germany (1–3). Physicians are confronted with patients suffering from diffuse symptoms that they are not able to explain by conventional differential diagnosis. In some cases, symptoms are like those of the sick building syndrome as described by Møhrave (4) and correlate clearly with contamination of xenobiotics at the place of employment. On the other hand, diffuse symptoms and undefined illness are related to air quality and existence of potentially harmful substances in homes. Lohmann (5) and Ohnsorge (6) have suggested that the use of wood preservatives in homes is a cause for neuropsychological and respiratory disorders. In 1990, Singer (7) described a number of similar neuropsychological symptoms, which are typical signs of chronic neurotoxicity. Wood preservatives used indoors in Germany contained mainly pentachlorophenol and hexachlorocyclohexane (lindane) until 1979; hexachlorocyclohexane, dichlorfluorane, and others since 1980; and mainly pyrethroids since 1986. These pesticides are known to be neurotoxic (8).

Because of an increased demand for experts in environmental medicine and the lack of possibilities of analysis of harmful substances in homes and workplaces, a cooperation was started between the Union of Physicians of Schleswig-Holstein (Bad Segeberg, Germany) and an environmental engineer in 1992. A mobile unit for environmental monitoring, sampling, and analysis was set up; this was called the Environmental Analysis Van (EAV). Inspection of sites and air and dust/material sampling for analysis of xenobiotics in homes or at the workplace could be performed on request.

Every physician who is a member of the Union of Physicians of Schleswig-Holstein can advise patients to request such an inspection or an environmental analysis of their homes or workplaces. The costs of sampling and analysis are covered entirely or partly by the patients' health insurance companies. Thus, if a physician suspects xenobiotics to be the cause of adverse health effects, she or he can advise environmental analysis. A report on the results of this unique cooperation in environmental medicine was considered noteworthy to show which sources of indoor pollutants are particularly relevant to human health impairment.

Methods

In the German Federal State of Schleswig-Holstein, physicians who suspect xenobiotics in the home or workplace to be the cause of symptoms of illness can advise analysis of the suspected environment. Upon call, the EAV inspects sites and, if necessary, takes samples of air and dust/material for analysis of xenobiotics. Samples of wood are collected with a specialized drill (depth, 2 mm; area, 10 cm²). Samples of dust are collected with a vacuum cleaner, and only a fraction of dust <63 μm is analyzed. For analysis of carpeting, a 10-cm² sample is taken and only the fiber fraction is analyzed. Samples of air are collected with an active sampler.

Analysis of biocides and volatile organic compounds (VOCs), as well as a few other xenobiotics rarely found (e.g., phthalates, asbestos, heavy metals, etc.), is done in cooperation with specialized laboratories. Because analytical methods and precision of analysis were not part of the database we examined, we do not discuss these items in detail, but the laboratories regularly participate in quality checks by analyzing external standards with unknown concentrations.

Consent is obtained from the client, and data from each inspected site is recorded by the environmental engineer operating the EAV on a specially designed data sheet. Data include work or living surroundings, housing characteristics, suspected xenobiotics, sampling material, source and amount of xenobiotics found, and/or symptoms of illness described by inhabitants or workers.

In July 1993 an evaluation of the data sheets was started at the Institute of Toxicology, University of Kiel, Germany. For this purpose, the data were made anonymous as required by data protection officials. For evaluation, the clients of the EAV were categorized as follows:

Category I. In this category, persons living or working at the examined site showed symptoms of illness (symptoms of illness correspond to the toxic effects of pollutants

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found in the homes or places of employment of the patients as described in the scientific literature), and one or more xenobiots were found in the immediate surroundings (homes or places of employment). The values found were above current threshold limit values allowed in Germany (if threshold limit values did not exist for the substance measured, an exposure was assumed if the value did exceed the background concentration in German homes (9–16)).

**Category II.** In Category II, persons living or working at the examined site showed symptoms of illness (symptoms of illness corresponded to the toxic effects of pollutants found in the homes or places of employment of the patients as described in the scientific literature) and one or more xenobiots were found in the immediate surroundings (homes or places of employment). The values found were just below current threshold limit values (if threshold limit values did not exist for the substance measured, an exposure was assumed if the value did exceed the background concentration in German homes (9–16)), particularly with reference to formaldehyde found in concentrations of 0.05–0.099 ppm, because the scientific literature describes adverse effects of formaldehyde below the German threshold limit value of 0.1 ppm (12).

**Category III.** This category includes persons living or working at the examined site who showed no symptoms of illness; analysis of environment was done for prevention only. One or more xenobiots were found in the immediate surroundings (homes or places of employment). The values found were above current threshold limit values (if threshold limit values did not exist for the substance measured, an exposure was assumed if the value did exceed the background concentration in German homes (9–16)); formaldehyde values ≥0.05 ppm were put into this preventive category.

**Category IV.** In category IV, persons living or working at the examined site showed symptoms of illness (symptoms of illness corresponded to the toxic effects of pollutants found in the homes or places of employment of the patients as described in the scientific literature). Environmental analysis of immediate surroundings (homes or place of employment) was performed, but values of xenobiots currently analyzed were below current threshold limit values allowed in Germany (if threshold limit values did not exist for the substance measured, an exposure was assumed if the value did exceed the background concentration in German homes (9–16)). The environmental engineer advised further testing, but no other tests were performed.

**Category V.** In this category, no xenobiots were found in the immediate surroundings (homes or places of employment) or the values found were below current threshold limit values allowed in Germany (if threshold limit values did not exist for the substance measured, an exposure was assumed if the value did exceed the background concentration in German homes (9–16)). Further testing was not necessary.

Because of a continuing discussion on possible adverse health effects of permethrin (a synthetic pyrethroid)-treated carpeting, a follow-up of persons exposed to permethrin was carried out. The period of time covered by the follow-up study was June 1994 until May 1995 (12 months). The results of this follow-up represent all cases in which elevated concentrations of permethrin were found because of carpeting in homes. All sites where other xenobiots were found in addition to pyrethroids were excluded from this follow-up. Similarly, all cases where pyrethroids were used for purposes other than carpet preservation were excluded.

After these exclusions, 83 clients of the EAV remained; the examined sites in this group had elevated concentrations of permethrin (>5 mg/kg) that had been found in dust or other materials because of the use of pyrethroid-treated carpeting. These clients were contacted by letter and again by telephone if they did not respond. Responses were received from 80 clients (96%). The clients were asked whether they removed one or all of the contaminated carpets. Furthermore, they were interviewed with regard to their health status and the period of time that had passed since the carpets were removed via a standardized questionnaire.

The results presented in this paper are only part of an overall documentation of activities in environmental medicine in Schleswig-Holstein. Additionally, data from patients who were treated by physicians qualified in environmental medicine have been reported (7). The relationship of environmental exposure and chronic neurotoxic diseases and multiple chemical sensitivity disorder has been reported by Lohmann et al. (18).

Data input and analysis were done with Epi Info (Version 6; Centers for Disease Control and Prevention, Atlanta, GA). Tables and figures were prepared with Microsoft Excel (Microsoft, Redmond, WA). Statistical analysis was done with Asuret (Statistics Add-In for Excel; DDU Software, Leeds, U.K.). The chi-square test was used for the assessment of the effect of removing pyrethroid-contaminated carpeting on the subjective health improvement of the participants in comparison with not removing them. Pawlik’s corrected contingency coefficient was calculated as described by Sachs (19).

**Results**

During a 30-month period from July 1993 to December 1995, 1793 site inspections were conducted by the EAIV. Xenobiotic analysis and subsequent advising was offered in 1318 instances. In 475 cases, no xenobiotic analysis was done or required, but inspection and subsequent advising was offered.

The results shown here include the 1318 cases in which environmental analysis was performed. Figure 1 illustrates the percentage of distribution of these 1318 cases in the categories I–V described above. In 71% of these cases, enhanced concentrations of one or more toxic substances were found (categories I, II, and III). In 21% of the cases, no enhanced concentrations of the analyzed substances were found (category IV). Further analysis was advised but not carried out until the end of December 1995. [For example, formaldehyde concentrations were analyzed in cases where there was new furniture, but the concentrations were found to be low; subsequent analysis of VOCs was advised because of the smell of the new furniture.] In 8% of cases, sufficient analysis of xenobiots was performed, but concentrations were low and no further suspicion of exposure to xenobiots arose (category V).

For the purpose of further analysis, only the data obtained in the categories I, II, and III were considered (n = 936). Figure 2 illustrates the percentage of xenobiots or classes of xenobiots (e.g., biocides) found in the 936 sites categorized as I, II, or III. Toxic substances mainly found in enhanced concentrations were biocides (n = 622) used for the preservation of wood (73%) or carpets (36%) or for pest control (2%).

![Figure 1. Percentage of clients of the Environmental Analysis Van who were placed in categories I–V between July 1993 and December 1995 (n = 1318).](image-url)
Predominantly the following substances were found: pentachlorophenol (PCP), permethrin, hexachlorocyclohexane (lindane), and dichlofluanid. Table 1 shows the concentrations of the main biocides found in homes where persons described symptoms of illness (category I, n = 512).

Enhanced concentrations of formaldehyde (≥20.05 ppm) were found in 311 cases (33%). Elevated concentrations (>1000 μg/m³) of VOCs were found in 6%; the results of the determination of single VOCs are not shown. Contamination with molds was found in 7% of cases. Other xenobiotics found in enhanced concentrations in less than 10 cases were phthalates, asbestos, polycyclic aromatic hydrocarbons, heavy metals, polychlorinated biphenyls, and hydrochloric acid.

In categories I, II, and III (n = 936), the sites of exposure were indoor living quarters (94%), cellars or attics (29%), places of employment (2%), and other sites or sources of exposure (2%; e.g., drinking water, a caravan, a church, etc.).

Symptoms of illness as described by the clients of the EAV were documented according to the kind of exposure (only pentachlorophenol, permethrin, and formaldehyde). The principal symptoms of illness described by persons solely exposed to ≥100 mg pentachlorophenol per kilogram of material (n = 69); with a main source of wood preservatives were respiratory problems (44%), tiredness (36%), and headaches (23%). The principal symptoms of illness described by persons solely exposed to formaldehyde (≥20.05 ppm; n = 173) were respiratory problems (58%), irritated eyes (36%), and headaches (31%). However, not all the symptoms can necessarily be attributed to indoor pollutants.

In the last few years, pyrethroids have been recognized as indoor xenobiotics bearing relevance to human health. Before 1994, indoor assessment of pyrethroids was rare. In the investigated period (30 months), exposure to pyrethroids above background levels (≥1 mg/kg dust, ≥5 mg/kg material) was found in 276 sites (categories I, II, and III). In 97% of these sites, permethrin was the pyrethroid measured at elevated concentrations (see Table 1). The maximum amount of permethrin found in dust was 5,000 mg/kg after the use of permethrin-containing pesticides for pest control; in most cases, however, the exposure was caused by pyrethroid-treated carpeting.

A follow-up of permethrin-exposed persons was carried out (see Methods for selection criteria). Eighty-three clients were contacted, and answers were obtained from 80 clients (96%). The clients were asked whether they removed one or all of the contaminated carpets. Furthermore, they were questioned with regard to their health status and the period of time that had passed since the carpets were removed. Seventy-five of the 80 contacted clients initially had called the EAV because of health impairments, whereas 5 clients had done this only for preventive reasons. The principal symptoms described by these persons (n = 75) who felt ill where permethrin was found were respiratory disorders (32%); burning eyes (32%); headaches (29%); dizziness (21%); tiredness (20%); pain of muscles, bones, and/or joints (20%); and skin effects (19%). Furthermore, symptoms such as tingling, burning, and/or numbness in the extremities were described by several exposed persons (13%). However, not all the described symptoms can necessarily be attributed to pyrethroids.

Table 2 shows the results of the follow-up. Forty-seven (59%) of the contacted clients had removed all carpet that was contaminated with permethrin. Eight (10%) clients had removed part of the contaminated carpet, and 25 (31%) clients did not remove any carpet. Thirty-nine (83%) of the clients who had removed all contaminated carpeting described a complete or at least a partial improvement of their complaints. The complaints of 6 (13%) clients did not improve in spite of removing their carpet. A minimum of 1 month since the carpeting had been removed had passed in all these cases, and in 81%, more than 3 months had passed.

Of the clients who did not remove any carpet, 44% did not recover from their symptoms of illness. The effect of removing all contaminated carpeting on health...
improvement in comparison with the results of not removing any carpet was statistically significant ($\chi^2; p<0.0001$). Pawlik’s corrected contingency coefficient was 0.71. The effect of removing all contaminated carpets in contrast to not removing any carpet is illustrated by Figure 3. The results suggest that the exposure to permethrin could indeed be the cause of adverse health effects.

The lowest concentrations of permethrin, which were assessed at the homes of clients who described complete recovery after removing all contaminated carpeting, were 10–15 mg/kg dust ($n = 4$) and 15 mg/kg carpet ($n = 1$).

In three cases where the contaminated carpets existed for 5, 7, and 10 years in the homes, permethrin concentrations were 115, 100, and 150 mg/kg dust, respectively. This result indicates that indoor permethrin contamination is highly persistent.

**Discussion**

Environmental medicine is a very difficult area of modern medicine. Forty thousand chemicals in more than 2 million formulations were known to be in industrial use, even in 1976 (7), and most of these xenobiotics have not been toxicologically evaluated. The practice of environmental medicine is a gigantic task in view of the number of xenobiotics that are possible causes of illness at places of employment or in homes. Many of these xenobiotics are potentially harmful to humans.

Usually, a limited number of chemicals is used in contaminated workplaces, and these substances are often known to the employees. The question of sick building syndrome can be elucidated by exploring the time of exposure and occurrence of symptoms and by asking how the patient feels over weekends and on vacation. An environmental engineer or industrial hygienist should then survey the site in question (21). The approach is more difficult if exposure to harmful substances happens in the home because knowledge of relevant indoor xenobiotics is limited and it may be a long time until the onset of illness. A detailed approach to diagnosis in environmental medicine is given by Cullen et al. (22).

The results of this report of the activities of an EAV did not arise from a controlled study, although a controlled follow-up was performed regarding one special kind of exposure to permethrin. It has been shown that in this small rural part of Germany (Schleswig-Holstein) xenobiotics or classes of xenobiotics relevant to human health are mainly pesticides used indoors (PCP, hexachlorocyclohexane, permethrin, dichlofluanid) as well as formaldehyde, VOCs, and molds. In only 3% of the inspected sites were other than the described substances or groups of substances found in elevated concentrations exceeding threshold limit values or background levels.

Although the intensity and frequency of exposure to xenobiotics relevant to human health may be different in urban and rural areas, other scientists (mainly from stationary environmental units or laboratories in big cities) report similar results (12,14,23).

Wood preservatives, which have been used in Germany in large amounts (in the range of 5 million homes), consisted mainly of pentachlorophenol and hexachlorocyclohexane until 1979, hexachlorocyclohexane and dichlofluanid and others since 1980, and mainly of pyrethrins since 1986 (20). All these substances are neurotoxic and highly persistent in treated wood. PCP was contaminated with polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs); these substances can still be found in the air surrounding treated wood more than 18 years later (24).

Cline et al. (25) found that PCP concentrations in people living in PCP-treated log homes were elevated in comparison with unexposed persons. Kerkvliet et al. (26) and Parker et al. (27) found enhanced concentrations of PCDDs and PCDFs in tissues of cattle exposed to PCP-treated wood or food, respectively. Kerkvliet et al. (26) and Thomas et al. (28) described health impairment of cattle kept on PCP-treated wood shavings or in PCP-treated barns, respectively. However, whether those biocide-containing wood preservatives are the cause of neurotoxic effects in humans is difficult to prove because the period of latency is very long and symptoms are unspecific, although often severe.

Lohmann et al. (17) documented that exposure to PCP-containing wood preservatives lasted 210 years in 68% of patients who developed symptoms of neurotoxicity. Only 19% of the patients described an onset of symptoms in the first 2 years of exposure. Singer (7) postulated that symptoms of chronic neurotoxicity caused by various substances are similar and include chronic fatigue, headaches, concentration difficulties, mental slowness, irritability, amotivation, social withdrawal, problems with memory, sexual dysfunction, and numbness of hands or feet. This has been confirmed in various case-control studies investigating the effects of chronic exposure to different neurotoxic organic solvents (29–34). Health impairment in connection with wood preservatives is described by various authors (6,17,35–40), whereas Liebl et al. (41) have not found any correlation between PCP values in serum and number and degree of symptoms of people living in log homes. But all these studies have serious limitations. Often, the authors did not consider confounders like age, gender, time, or end of exposure or existence of other neurotoxic substances in the homes or places of employment (hexachlorocyclohexane, PCDDs/PCDFs, pyrethroids, organic solvents, etc.).

Because PCP has been prohibited in Germany since 1989 and has generally not been used indoors since 1979, PCP seemed to be eliminated as an indoor contaminant. However, in the present report, PCP is found in elevated concentrations in 231 homes where people have described health impairment. Because technical PCP is contaminated by PCDDs and PCDFs, elevated PCP values in wood and dust indicate an exposure to PCDDs and PCDFs. Furthermore, we must be aware that times of exposure before illness may be very long. Therefore, PCP continues to be an indoor contaminant and continues to impair human health.

Pyrethroids have gradually replaced PCP, hexachlorocyclohexane, and other biocidal substances in all kinds of pesticide formulations since 1980. In Germany, more than 50% of pesticides used indoors contain pyrethroids. They are used in wood preservatives and for pest control (in insect sprays and powders, in insecticide vaporizers, and in wool carpets to prevent damage from pests such as moths or beetles) (14). Schulz et al. (15) have found that, in Germany, 31% of a representative group of persons (25–69 years old, $n = 4021$) use some kind of biocide indoors. Twenty percent use wood preservatives and 26% use insecticides.

Synthetic pyrethroids, which are mainly used, are neurotoxic. They have very low vapor pressure and show high adsorption to...
very quickly after use of pyrethroid-containing spray. At the same time concentrations in surrounding objects and dust will rise quickly. Pyrethroids can be inhaled and absorbed dermally by persons living in contaminated rooms (14,15). In rooms where there are pyrethroid-treated carpets, these pyrethroids can be found in dust as well as on the surface of furniture (14). In Germany the main pyrethroid used indoors is permethrin. The background levels of permethrin in German homes are <1mg/kg dust and <5mg/kg material (13).

In a 30-month period, the EAV found concentrations of permethrin and other pyrethroids in 276 sites, and these concentrations exceeded the background levels by sometimes a hundredfold. The maximum amount of pyrethroids found in dust was 5,000 mg permethrin per kilogram after use of pesticides for pest control. However, in 83% of sites, the source of pyrethroids found by the EAV was carpeting.

Although acute intoxication with pyrethroids is described by several authors (42–46), the potential of adverse health effects on adults and children after low chronic indoor exposure to this neurotoxic substance is disputed (14,17,43,47,48). Corrigan et al. (49) described a case of persistent illness after occupational exposure to permethrin. In a preliminary investigation, Fiedler et al. (50) reported that Gulf War veterans, who have been exposed to pyrethroids and other pesticides, display symptoms characterized by severe fatigue and multiple chemical sensitivities. Aboudonia [see (51)] confirms this relationship; he describes neurotoxic symptoms in Gulf War veterans as characterized by chronic fatigue, rashes, headaches, weight loss, and joint pain and suggests that synergy between different pesticides may be responsible for these effects.

Lohmann et al. (17) documented the onset of illness in 21 patients with mainly chronic pyrethroid exposure and reported that symptoms began in 62% of these patients in the first 2 years of exposure. Thus, the latency between exposure to pyrethroids and occurrence of symptoms seems to be much shorter than the corresponding latency after exposure to PCP. Müller-Mohnssen and Hahn (43) described symptoms of chronic exposure to pyrethroids, which are similar to the symptoms postulated by Singer (7) for chronic neurotoxicity. Müller-Mohnssen and Hahn (43) described severe cases of pyrethroid exposure in which muscle pain, uncertain gait, dizziness, numbness, burning, and/or feelings of pins and needles in the extremities were also observed; this has been confirmed by Lohmann et al. (17). But these authors only describe their observations of cases so the evidential value is therefore limited. Similar symptoms have been described by approximately 10–30% of 75 clients of the EAV with elevated concentrations of pyrethroids and who were included in the follow-up study. Altenkirch and Hopmann (48), in a case study of 26 patients with supposed pyrethroid exposure, did not find a correlation between exposure and health impairment, but this study is highly criticizing (47,52,53). Furthermore, pyrethrum and some synthetic pyrethroids have been discussed as potential allergens (44–47, 54–57). A controlled study should be carried out to investigate the health effects of chronic exposure to pyrethroids indoors.

The results of the follow-up performed on 80 clients of the EAV who had been exposed to permethrin indicate that indeed permethrin-treated carpeting can be the cause of adverse health effects. Eighty-three percent of the clients who did remove all contaminated carpeting described complete or at least partial recovery. Although it is possible that this is a placebo effect, it is not likely. It could be argued that the removal of carpets would have health benefits anyway, as overall dust and mite levels would be reduced significantly. On the other hand, clients of the EAV frequently report that symptoms of illness began in the time after they bought new carpeting containing permethrin. Most of them had carpeting before, as do about 90% of German households. This question and others concerning exact health status before exposure, including allergies, dust levels, type of carpeting used before, etc., should be investigated further using another study design (exposed vs. not exposed).

At three sites where the contaminated carpeting already existed for 5, 7, and 10 years in the homes of clients of the EAV, there were still permethrin concentrations of 115, 100, and 150 mg per kilogram of dust. This result indicates that indoor contamination of permethrin is highly persistent and also confirms the findings of Stolz (58), who reported that the half-life of long-lasting pyrethroids (permethrin, deltamethrin, cypermethrin, cyfluthrin) is about 10 years indoors.

The lowest concentrations of permethrin, which were assessed in the homes of clients who described complete recovery after removing all contaminated carpeting, are 10–15 mg/kg dust (n = 4) and 15 mg/kg carpet (n = 1). Taking into consideration that these concentrations are only about 10 times above the alleged background level in German homes and regarding the high persistence of pyrethroids indoors, we must wonder whether the indoor use of pyrethroids is safe enough to avoid adverse health effects.

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