

German Environmental Survey: Exposure from indoor air and indoor dust

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The German Environmental Survey (GerES) is a nationwide population study which has repeatedly been carried out in Germany since the mid-1980s [1]. The first survey for adults (GerES I) was carried out in 1985/1986 (West Germany) followed by GerES IIa in 1990/91 (West Germany) and GerES IIb in 1991/92 (East Germany). In GerES II children were included to some extent [2, 3]. In 1998, the third GerES for adults was conducted in both parts of Germany (GerES III, [4]). The 2003/2006 survey (GerES IV) focussed exclusively on children [5]. It was preceded by a one-year pilot study (2001 to 2002) to collect information on parameters influencing the response rate and to test the different instruments intended to be used for the main study. All GerESs were conducted in cooperation with a respective German Health Survey. GerES IV for example was conducted in cooperation with the National Health Interview and Examination Survey for Children and Adolescents (KiGGS) carried out by the Robert Koch Institute, Berlin [6].

The main goals of GerES are to analyse and document the extent, distribution and determinants of exposure to environmental pollutants of the general German population. Three main instruments of investigation were comprised in GerES: human biomonitoring, monitoring of the domestic environment, collecting data on exposure factors via questionnaires as well as the measurement of noise and detection of hearing impairments. Monitoring the domestic environment is vital to detect indoor exposure pathways and to quantify their contribution to overall exposure.

Materials and methods

GerES is a cross-sectional study. In GerES IV 1,790 children aged 3 to 14 years were selected at random from the representative KiGGS sample. The domestic environment of about 600 randomly selected children was monitored, *inter alia*, by analysing Volatile Organic Compounds (VOC) in indoor air and different pollutants in indoor dust [1, 5]. For the GerES IV Pilot Study which was carried out between March 2001 and March 2002, 550 children aged 0 to 17 years were selected via municipal inhabitant registers or schools in 4 sampling locations (two districts in the city of Berlin and two in rural areas in other federal states [7]).

In GerES IV the VOC were usually determined in the children's bedroom. Samplers were placed in an appropriate place and remained there for approximately one week. Non-polar VOC were determined by using 3M OVM-3500 samplers. Polar and high-boiling VOC were measured in addition to non-polar VOC by a tube type sampler (Perkin Elmer). SKC UMEx 100 badge type samplers were used for formaldehyde and other carbonyls [8]. Analysis of the Perkin Elmer samplers and determination of the specific uptake rates were performed at Ergo-Research in Hamburg, a laboratory that holds accreditation for these analyses. The analyses of the UMEx 100 and the OVM 3500 samplers as well as additional quality control measurements were carried out in the laboratories of the Federal Environment Agency. Sampling and analyses of VOC in GerES I (1985/86) were described by Seifert *et al.* [9]. Sampling and analytical methods used in the two surveys are comparable.

To collect household dust, the vacuum cleaner bags as found in the home at the time of the visit were used. After collection in the home each vacuum cleaner bag was placed in a plastic bag for transportation. In the laboratory the content of the bag was sieved (63 µm fraction) and filled in glass jars. All materials used were checked for relevant blanks. To determine phthalates the dust samples were extracted with toluene and measured with GC/MS or HPLC/MS [10].

The analytical procedure to determine the metabolites of phthalates like MnBP, MBzP, MEHP, 5OH-MEHP and 5oxo-MEHP in morning urine was described elsewhere [11, 12].

All statistical calculations were performed using SPSS 14.0 for MS Windows. Concerning VOC values below LOQ were set to 2/3 LOQ for data treatment according to Hallez and Derouane [13]. Concerning dust, values below LOQ were set to LOQ/2.

Results and discussion

According to questionnaire data from GerES IV children aged 3 to 14 years on average spent between 20 and 21 hours daily in indoor environments, most of the time at home. Thus a clean environment at home is very important for the children's health. In the following some selected results of GerES concerning indoor exposure are presented to make these results accessible to the scientific community on short call.

In Figure 1 some of the VOC concentrations measured in indoor air in GerES IV are compared with the concentrations quantified in the former GerES I which was conducted in 1985/86 [9]. The geometric mean concentrations of aromatic compounds like benzene and toluene have decreased from 6.2 µg/m³ to 1.9 µg/m³ and from 50.7 µg/m³ to 13.6 µg/m³, respectively. The same applies for alkanes, here shown for n-decane, and for chlorobenzenes, here shown for 1,4-dichlorobenzene.

It has to be mentioned that in GerES I the measurement of the VOC took place in the living rooms of

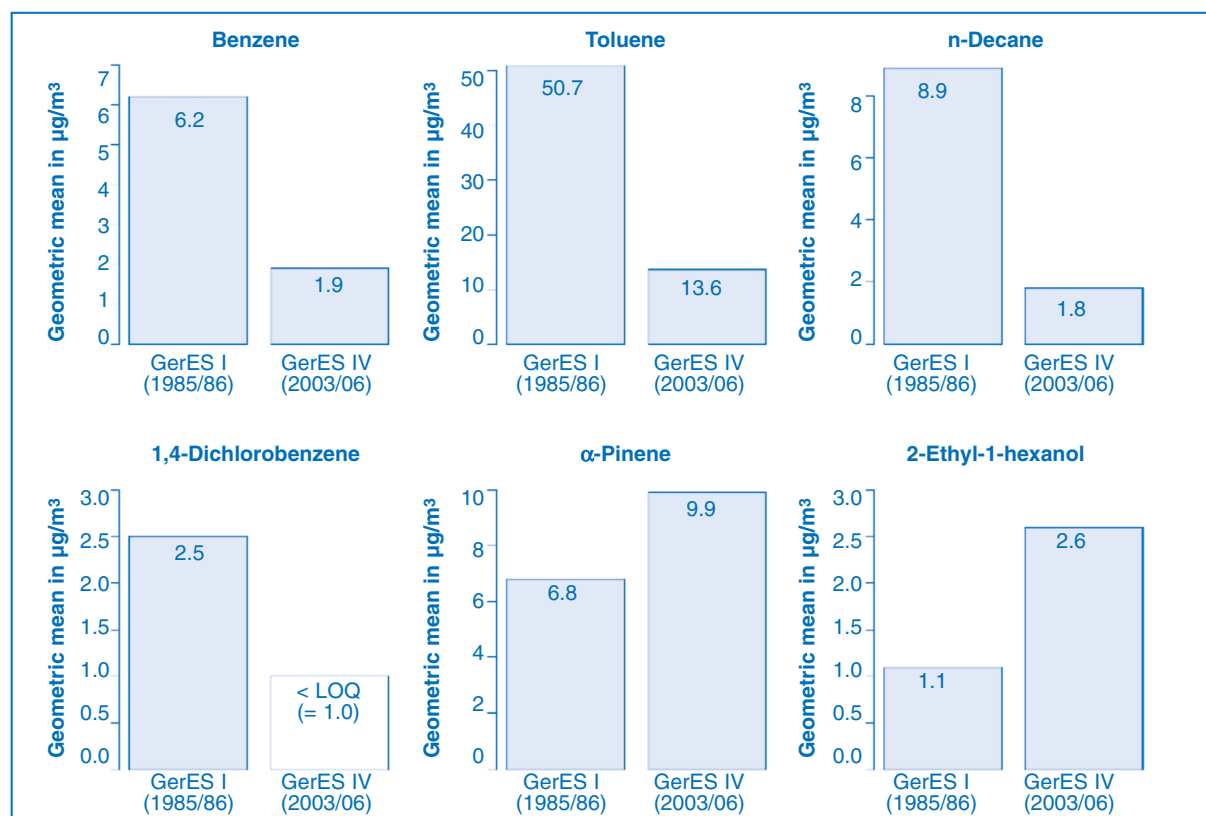


Figure 1.
Time trend of selected VOC in indoor air in German homes.

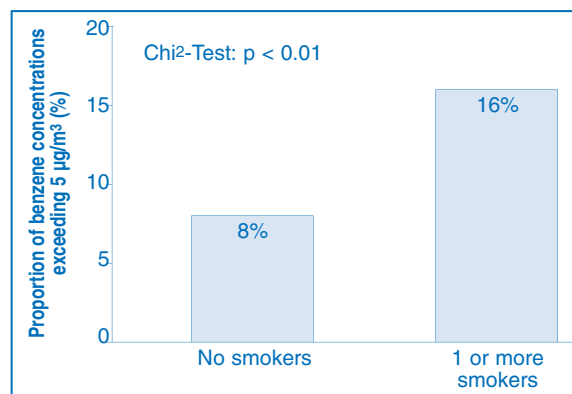


Figure 2.
Benzene in indoor air of smokers and non-smokers in German homes (percentages exceeding 5 µg/m³).

adults, regardless if children were present in the household or not. In GerES IV the samplers were generally placed in the bedroom of the child. Schneider *et al.* found lower VOC concentrations (except for benzene) in adult's bedrooms than in living rooms or kitchens [14]. However, the time trends shown in the examples are very clear and unlikely to be confounded substantially by the difference in sampling place.

The reason for the decreasing concentration of benzene is the reduction of aromatic hydrocarbons in urban air due to the introduction of automobile cata-

lytic converters in the last two decades and the reduction of the benzene content of gasoline. The decrease of toluene concentration can be ascribed to the ongoing reduction of toluene containing solvents in building and household products. However, high concentrations can still be observed. Since a limit value for benzene in indoor air is not available the future EU outdoor air threshold value of 5 µg/m³ might be appropriate to assess the results of GerES IV. Figure 2 shows that in homes with one or more smokers 16% of the measured benzene values exceed this value compared to 8% in homes with no smoker. Questionnaire data from GerES revealed that still around 50% of the children lived in households with at least one smoker.

N-decane (Figure 1), like other n-alkanes is a component of cleaning products, oil, wax and paints. 1,4-dichlorobenzene is a biocide used for example in mothballs. High concentrations of α -pinene can be expected if new wooden furniture is present in a home [9]. It might also be possible that wooden "natural" or "organic" products which became fashionable in Germany in recent years are sources of pinenes. 2-Ethyl-1-hexanol is a breakdown product of the phthalate plasticizer DEHP.

Exposure from the indoor environment at home might not only occur from indoor air but also from house dust. Therefore in all GerES conducted so far the examination of house dust was included.

Table 1.
Phthalates in house dust (mg/kg, 63 µm-fraction, [10]).

Phthalate	P50	P95
BBzP	13	180.0
DnBP	29	180.0
DEHP	480	1,700

Note: Results from GerES Pilot Study (N = 278), P50 = median, P95 = 95th percentile.

Table 2.
Correlations between phthalates in house dust and phthalate metabolites in urine of children.

	MBzP in urine	MnBP in urine	MEHP in urine	5OH-MEHP in urine	5oxo-MEHP in urine
BBzP	0.11	---	---	---	---
DnBP	---	0.23 *	---	---	---
DEHP	---	---	0.06	0.00	0.00

Note: Results from GerES Pilot Study (N = 254), *: p < 0.001, Spearman rank correlation.

In Table 1 the results for some of the phthalates analysed in the GerES IV Pilot Study are shown: BBzP (butylbenzylphthalate), DnBP (Di-n-butylphthalate) and DEHP (diethylhexylphthalate, [10]). DEHP was present in high concentrations with a 95th percentile of 1,700 mg/kg. BBzP and DnBP both showed a 95th percentile of 180 mg/kg. In the GerES IV Pilot Study the metabolites of these phthalates had been analysed in the urine of the participating children [7, 12]. It was an inviting task to correlate the concentrations measured in house dust with the concentrations of the phthalate metabolites measured in urine.

The results are shown in Table 2. There was a significant correlation (p < 0.001) between DnBP in house dust and the DnBP metabolite MnBP (mono-butylphthalate) concentration in urine. Compared to BBzP and DEHP, DnBP has a higher vapour pressure which might cause a higher mobility of DnBP in the indoor environment and a corresponding transfer to humans.

Conclusion

The German Environmental Survey (GerES) is a worldwide unique example for successful exposure assessment of the general population using a combination of human biomonitoring and monitoring of the indoor environment at home in a large scale cross-sectional study. A detailed description of results on the VOC analysis in GerES IV will be published soon [15]. Analyses of phthalates in house dust and their metabolites in urine in GerES IV will substantiate the results presented.

Acknowledgement

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Références

- [1] Schulz C, Conrad A, Becker K, Kolossa-Gehring M, Seiwert M, Seifert B. Twenty years of the German Environmental Survey (GerES), Human biomonitoring – temporal and spatial (West Germany/East Germany) differences in population exposure. *Int J Hygiene Environ Health* 2007 ; 210 : 271-97.
- [2] Seifert B, Becker K, Hoffmann K, Krause C, Schulz C. The German Environmental Survey 1990/92 (GerES II): A Representative Population Study. *J Exp Anal Environ Epidemiology* 2000a ; 10 : 103-14.
- [3] Seifert B, Becker K, Helm D, Krause C, Schulz C, Seiwert M. The German Environmental Survey 1990/92 (GerES II): Reference Concentrations of Selected Environmental Pollutants in Blood, Urine, Hair, House Dust, Drinking Water, and Indoor Air. *J Exp Anal Environ Epidemiology* 2000b ; 10 : 552-65.
- [4] Becker K, Kaus S, Krause C, Lepom P, Schulz C, Seiwert M, Seifert B. German Environmental Survey 1998 (GerES III): Environmental pollutants in blood of the German population. *Int J Hyg Environ Health* 2002 ; 205 : 297-308.
- [5] Becker K, Schulz C, Babisch W, Dürkop J, Roskamp E, Seiwert M, Szewzyk R, Ullrich D, Seifert B. German environmental survey for children (GerES IV) 2003-2006. *Pollution Atmosphérique* 2005 ; 188 : 475-9.
- [6] RKI (Robert Koch Institute): KiGGS – The German Health Survey for Children and Adolescents. Berlin, Germany, 2006. Download: <http://www.kiggs.de>
- [7] Becker K, Seiwert M, Angerer J, Heger W, Koch HM, Nagorka R, Roskamp E, Schlüter C, Seifert B, Ullrich D. DEHP metabolites in urine of children and DEHP in house dust. *Int J Hyg Environ Health* 2004 ; 207 : 409-17.
- [8] Ullrich D, Gleue C, Krause C, Lusansky C, Nagel R, Schulz C, Seifert B. German Environmental Survey of Children and Teenagers 2000 (GerES IV): A representative population study including indoor air pollutants. In: *INDOOR AIR '2002* ; Vol. I. : 209-13.
- [9] Seifert B, Mailahn W, Schulz C, Ullrich D. Seasonal Variation of Concentrations of Volatile Organic Compounds in Selected German Homes. *Environment International* 1989 ; 15 : 397-408.
- [10] Nagorka R, Scheller C, Ullrich D. Weichmacher im Hausstaub. *Gefahrstoffe-Reinhaltung der Luft* 2005 ; 65 (3) : 99-105.
- [11] Koch HM, Gonzales-Reche LM, Angerer J. On-line cleanup by multidimensional LC-ESI-MS/MS for high throughput quantification of primary and secondary phthalate metabolites in human urine. *J Chromatogr B, Analyt Technol Biomed Life Sci* 2003 ; 784 : 169-82.
- [12] Koch HM, Becker K, Wittassek M, Seiwert M, Angerer J, Kolossa-Gehring M. Di-n-butylphthalate and Butylbenzylphthalate (BBzP) – urinary metabolite levels and estimated daily intakes: pilot study for the German Environmental Survey on children (GerES IV). *J Exposure Sc Environ Epidemiol* 2007 ; 17 : 378-87.
- [13] Hallez S, Derouane A. Nouvelle méthode de traitement de séries de données tronquées dans l'étude de la pollution atmosphérique. *Science Total Environ*. 1982 ; 22 : 115-23.
- [14] Schneider P, Lörinchi G, Gebefügi IL, Heinrich J, Kettrup A, Wichmann HE. Vertical and horizontal variability of volatile organic compounds in homes in Eastern Germany. *J Exp Anal Environ Epidemiol* 1999 ; 9 : 282-92.
- [15] UBA (Umweltbundesamt): German Environmental Surveys. Federal Environment Agency, Dessau/Berlin, July 2007. <http://www.umweltbundesamt.de/survey-e/index.htm>