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COMMUNICATION

Occurrence of organochlorine pesticides in indoor dust†

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Organochlorine pesticides are present in the environment and suspected of causing serious health effects. Diet has been the main exposure source, but indoor source release is gaining focus. Within a monitoring study of polychlorinated biphenyls of Danish buildings built during the 1960s and 1970s, we coincidently determined extreme levels of dichlorodiphenyltrichloroethane (DDT) levels in two of ten random samples. This raises concern and further large scale investigations are warranted to confirm this.

Persistent and lipid-soluble organochlorine pesticides such as dichlorodiphenyltrichloroethane (DDT) are ubiquitously present as a complex mixture of parent compounds and metabolites in the environment. DDT is a manufactured chemical, which was introduced in the 1940s and became one of the most widely used chemicals for controlling pests that are detrimental to agricultural crops and carry diseases such as malaria and typhus. In Denmark, DDT was restricted in 1969 and banned in 1986, but more than 520 metric tonnes of DDT have been sold in Denmark alone.[1] The organochlorines hexachlorobenzene (HCB), hexachlorocyclohexane (HCH) and chlordane were less extensively used in Denmark.

Although these compounds have been banned in most Western countries for decades, their previous extensive worldwide application, continued use in developing countries and environmental persistence[2] have resulted in continued redistribution in the environment. DDT, HCB and chlordane are regulated by the Stockholm Convention on Persistent Organic Pollutants (POPs), with the objective for parties to eliminate and reduce the emissions of POPs. HCH is included in the Aarhus Protocol on POPs adopted by the United Nations Economic Commission for Europe, 1998, with the ultimate objective to eliminate any discharge, emissions and losses of POPs.

Organochlorine pesticides are carcinogenic in animals[3] and their immunotoxicity has been documented *in vitro*,[4] *in ovo*,[5] as well as in animals[6–8] and human foetal, neonatal and infant immune systems.[9–11] Concern about health risks has prompted research into potential exposure sources. As the compounds bioaccumulate and biomagnify in the food chain, organochlorine pesticides have been detected in high concentrations in meat, milk and fish,[12–16] but lower levels in fruit, vegetables and cereals.[17] The indoor environment has been shown to be important for human exposure to several halogenated organic pollutants such as polychlorinated biphenyls[18] and brominated flame retardants.[19] However little is known about indoor sources of organochlorine pesticides. Prior to its ban, the direct application of DDT to treat new carpets and textiles for pests was a common practice.[20] Carpet and other textiles act as long-term reservoirs for organochlorine pesticides,[21–23] which are continually transferred to indoor air and dust.[24,25] Organochlorine pesticides bound to dust are more persistent indoors due to a lack of degradation (microbial and photolytical) and other dissipation (volatilisation, dissolution) processes.[25,26] The fact that populations in industrialised countries spend 85–90% of their time in indoor environments[27] increases the relevance of indoor exposure to these compounds. Quantification of the amount of organochlorine pesticides in indoor environments is presently the focus of increasing concern and four recent studies have reported elevated concentrations in indoor dust[21,23,25,28] in industrialised countries, whilst two others have related these concentrations to cancer.[22,29]

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Environmental impact

Lipid-soluble organochlorine pesticides are ubiquitously present in the environment and have been detected worldwide in humans. These compounds are of environmental health interest because of their persistent nature and potential adverse health effects. Established human exposure sources include meat, milk and fish, but there is presently increasing focus on the indoor environment. Although several research groups have investigated the indoor environment in relation to organochlorines such as PCBs and PBDEs, little is known about the indoor environment as a source of exposure to organochlorine pesticides such as DDT. This is one of the first reports indicating that indoor dust is an important DDT exposure source and this raises concern—not least for children's health—and addresses the ongoing debate about human exposure sources of organochlorines.

In the present study we report the concentrations of DDTs, α-, β- and γ-HCH, HCB, and *trans*-nonachlor (TNC) in household dust collected from ten Danish addresses.

Between October 2003 and December 2003, municipalities in the Eastern Zealand of Denmark and the National Knowledge Center for Caulking were invited to provide possible locations for the study. Questionnaires were then sent out to the owners of 100 selected indoor localities to gather information on potential sources of polychlorinated biphenyls. Based on the information in these questionnaire replies, ten of these addresses (Fig. 1) were selected for chemical analyses of window sealants, indoor air, indoor dust and garden soils. Localities were selected based on participant willingness, suspicion of PCB contamination and proximity to the Danish building Research Institute in Hoersholm. The ten localities represent the different municipalities along the Eastern Zealand coast of Denmark (see Fig. 1) and also represent varying building types including six residential (five private one-family homes and one apartment) and four educational (two schools and two university research facilities) buildings. All of the selected buildings were constructed in the period 1960 to 1973, which corresponds to the period of the most intensive PCB usage in various types of sealants and also to the most intensive DDT usage worldwide (Table 1).

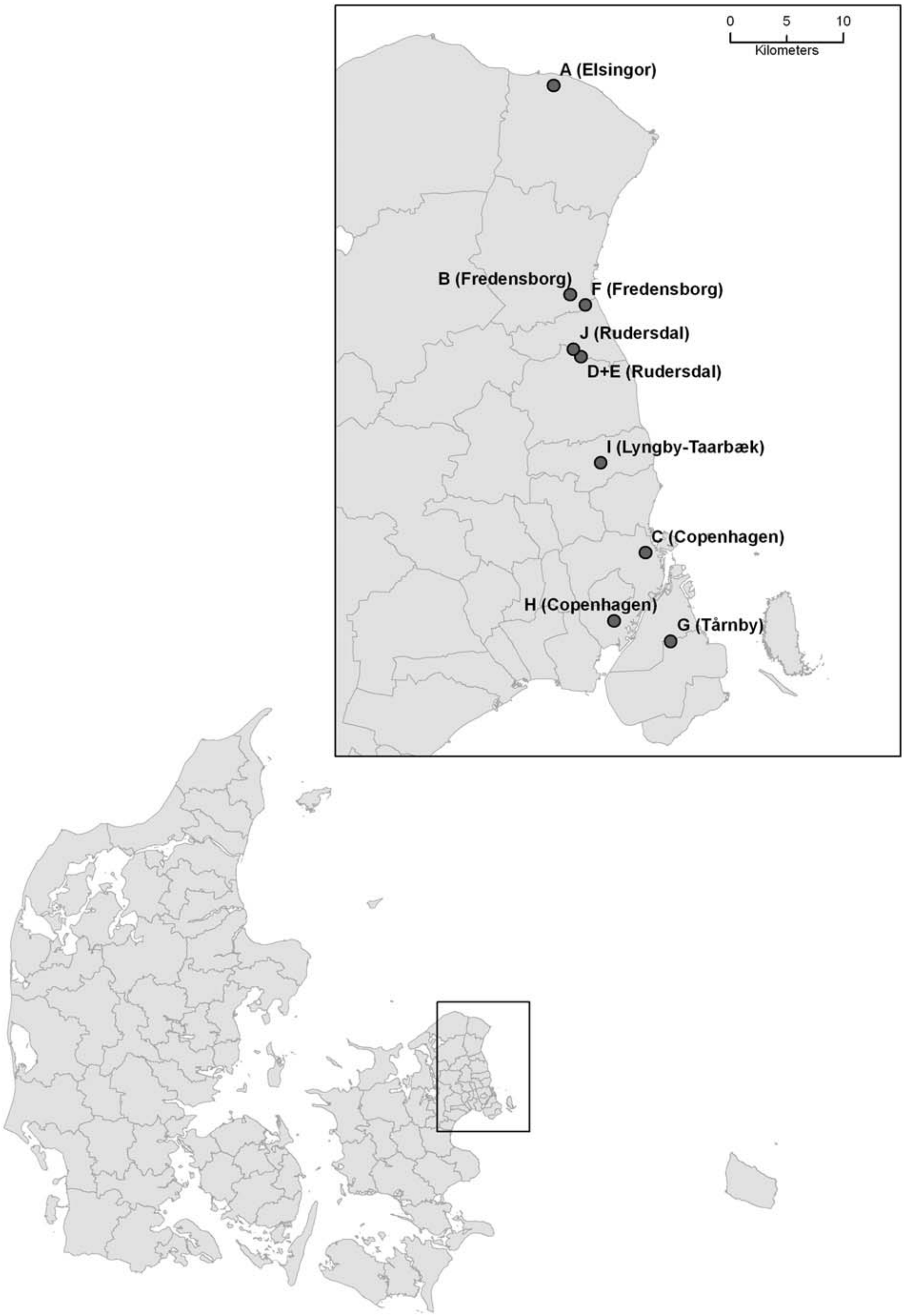

Fig. 1 Distribution of geocoded addresses ($n = 10$) in six Eastern Danish municipalities.

In January and February 2004 all questionnaire responses were reviewed and the measurements in the selected ten indoor localities were prepared. Sampling of indoor dust was undertaken between March 2004 and June 2004. A specially made filter cassette was mounted on a traditional vacuum cleaner (1600 W) for dust sampling. Dust was collected on circular glass fibre filters with a diameter of 80 mm. The suction opening was 10 mm × 60 mm. To ensure that the vacuum cleaner did not scrape the surface of the floor small wheels were mounted on the opening to maintain a distance of 5 mm between the vacuum cleaner head and the sampling surface (Fig. 2). A floor or carpet area of 2 m^2 was sampled during 2 minutes at each location. One sample was taken at each locality in front of a window in the most frequently used area at each location. The filters were immediately wrapped in aluminium foil and stored in sealed plastic bags for chemical analysis.

Details of organochlorine pesticide analyses in dust and gas-chromatography-spectrometry parameters are provided in our ESI†.

The dust concentrations (ng g^{-1}) of the organochlorine pesticides found in each of the ten localities are presented in Table 2. All of the DDTs, with the exception of *o,p*-DDE were extremely elevated in samples from private homes A and B, with respective levels of 1253 and 19 756 ng g^{-1} for ΣDDT. All other pesticides were detected in the dust samples from schools, institutions and the remaining private homes at relatively low levels or levels below the limit of detection (Table 2).

It is surprising that DDE, the major metabolite of DDT, was not recovered in samples A and B. But this is probably because ΣDDT bound to dust is more persistent indoors due to a lack of degradation and other dissipation processes.[25,26] Organochlorine pesticides are trapped in carpet and other textiles.[21–23] Interestingly, both samples with high levels of ΣDDT originated from private homes with carpeting on floors, whilst only one of the localities with low levels of ΣDDT had carpeting on floors. This supports the theory that carpets can act as a reservoir for ΣDDT, perhaps with an origin from the prior ban direct application of DDT for pest control or imported carpeting from lands still using DDT.

These high levels of ΣDDT raise concern. When considering oral exposure, the acute oral minimum risk level (MRL) for DDTs in humans is 0.5 μg^{-1} kg^{-1} day^{-1}.[26] Toddlers and younger children have vulnerabilities to chemicals that are distinct from adults. Their exposure is expected to be higher because of their activities on the floor and their tendency for ingestion of non-food items and hand-to-mouth behaviour.[30] The ingestion "central tendency" value of indoor dust in toddlers aged 1 to 3 years is 60 mg per day.[31] If we consider a 10 kg toddler in either private home A or B, then the ingestion of 60 mg of dust would be equivalent to a DDT intake of 0.0075 and 0.1186 μg^{-1} kg^{-1} day^{-1} respectively. These levels contribute significantly to the acute MRL for the average toddler and would be even greater for younger infants with lower body mass.

Fig. 2 A specially made filter cassette was mounted on a traditional vacuum cleaner (1600 W) for dust sampling. Small wheels were mounted on the opening to maintain a distance of 5 mm between the vacuum cleaner head and the sampling surface.

Concentrations of organic contaminants in dust are known to vary both spatially and temporally, and relative standard deviations of up to 200% have been shown for polybrominated diphenylethers congeners in indoor dust.[32] Although we did not include follow-up measurements and the absolute ΣDDT concentration could vary with time and within the room, there is a strong indication of significant DDT sources in private homes A and B.

The present results are based on ten indoor environments and may thus not reflect the true frequency of high DDT concentrations in dust inside all buildings in Denmark so we cannot draw any firm conclusions here regarding the indoor environment (dust, carpets, and textiles) as a source of DDT exposure or health risks at the population level. But these results are disquieting and need to be confirmed. Work on a larger scale, with the inclusion of indoor environments occupied by susceptible groups such as small children[30] in child day care centres and a wider range of persistent organic pollutants, is warranted.

In conclusion, the exposure to organochlorine pesticides has generally been associated with fatty fish and other food items, but

Table 1 Building characteristics

ID	Type	Year[a]	Area[a]/m^2	Carpet (Y/N)	Dust sample mass/g
A	Private home	1971	135	Y	1.330
B	Private home	1973	173	Y	0.055
C	Apartment	1960	137	N	0.054
D	Private home[b]	1961	80	N	0.058
E	Private home[b]	1961	115	N	0.048
F	Private home	1972	123	Y	0.092
G	High School	1969	870	N	0.103
H	High School	1972	2000	N	0.115
I	University, research centre	1963	4721	N	0.241
J	University, research centre	1973	10 318	N	0.191

[a] Building details obtained from the Danish Enterprise and construction Authority. [b] Private homes D and E were neighbours.

Table 2 Concentrations (ng g^{-1}) of the organochlorine pesticide in household dust from ten different localities[a]

	Concentration (ng g^{-1})									
	A	B	C	D	E	F	G	H	I	J
p′p-DDT	999	19 033	<15	17	76	68	47	17	9	42
o′p-DDT	95	201	<15	<15	16	18	14	<10	<5	6
p′p-DDE	86	100	<15	<15	<15	15	<10	<10	<5	<5
o′p-DDE	6	<15	<15	<15	<15	<10	<10	<26	5	24
p′p′-DDD	67	422	<15	<15	<15	<10	<10	<10	<5	<5
∑DDTs	1253	19 756	—	17	92	101	61	17	14	72
α-HCH	<1	<15	<15	<15	<15	<10	<10	<10	<5	<5
β-HCH	2	<15	<15	<15	<15	<10	<10	<10	<5	<5
γ -HCH	9	<30	<30	<30	<30	<20	<20	<20	<10	<10
HCB	<1	<1	3	<1	1	<10	<10	<10	<5	<5
TNC	<1	<15	<15	<15	<15	<10	<10	<10	<5	<5

[a] All sampling was undertaken between March 10 to June 29, 2004. Dust was sampled over a 2 m^2 area using a 1600 Watt vacuum cleaner for 2 minutes.

little attention has been paid to the indoor environment. This is one of the first studies reporting indoor dust as an important human exposure source of these compounds and the presence of high concentrations of DDTs in the indoor environment raises concern for human health. More work on a larger scale is warranted.

Abbreviations

DDT	dichlorodiphenyltrichloroethane
HCH	hexachlorocyclohexane
HCB	hexachlorobenzene
MRL	minimum risk level
PCB	polychlorinated biphenyl
POP	Persistent Organic Pollutant
TNC	*trans*-nonachlor

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References

1 T & A (Teknik & Administration) Nr. 4, Branchbeskrivelse for skovbruget med fokus på anvendelse af DDT, Amternes Videncenter for Jordforurening, Copenhagen, Denmark (in Danish), 1988.

2 ATSDR, *Selected PCBs (Acrochlor-1260, -1254, -1248, -1242, -1232, -1221 and -1016)*, Agency for Toxic Substances and Disease Registry, Atlanta, GA, USA, 1993.

3 International Agency for Research on Cancer, *Overall Evaluations of Carcinogenicity: An Updating of IARC Monographs Volumes 1 to 42*, International Agency for Research on Cancer, Lyon, France, 1987.

4 H. Bilrha, R. Roy and B. Moreau, et al., *In vitro* activation of cord blood mononuclear cells and cytokine production in a remote coastal population exposed to organochlorines and methyl mercury, *Environ. Health Perspect.*, 2003, **111**, 1952–1957.

5 E. T. Lavoie and K. A. Grasman, Effects of *in ovo* exposure to PCBs 126 and 77 on mortality, deformities and post-hatch immune function in chickens, *J. Toxicol. Environ. Health, Part A*, 2007, **70**, 547–558.

6 H. Tryphonas, M. I. Luster and K. L. White, Jr., et al., Effects of PCB (Aroclor 1254) on non-specific immune parameters in rhesus (*Macaca mulatta*) monkeys, *Int. J. Immunopharmacol.*, 1991, **13**, 639–648.

7 K. A. Grasman and G. A. Fox, Associations between altered immune function and organochlorine contamination in young Caspian terns (*Sterna caspia*) from Lake Huron, 1997–1999, *Ecotoxicology*, 2001, **10**, 101–114.

8 A. Bernhoft, J. U. Skaare and O. Wiig, et al., Possible immunotoxic effects of organochlorines in polar bears (*Ursus maritimus*) at Svalbard, *J. Toxicol. Environ. Health, Part A*, 2000, **59**, 561–574.

9 E. Dewailly, P. Ayotte and S. Bruneau, et al., Susceptibility to infections and immune status in Inuit infants exposed to organochlorines, *Environ. Health Perspect.*, 2000, **108**, 205–211.

10 C. Heilmann, P. Grandjean and P. Weihe, et al., Reduced antibody responses to vaccinations in children exposed to polychlorinated biphenyls, *PLoS Med.*, 2006, **3**, e311, DOI: 10.1371/journal.pmed.0030311.

11 R. Repetto and S. S. Baliga, Pesticides and immunosuppression: the risks to public health, *Health Policy Plan.*, 1997, **12**, 97–106.

12 M. P. Longnecker, W. J. Rogan and G. Lucier, The human health effects of DDT (dichlorodiphenyltrichloroethane) and PCBS (polychlorinated biphenyls) and an overview of organochlorines in public health, *Annu. Rev. Public Health*, 1997, **18**, 211–244.

13 WHO, *Health Risks of Persistent Organic Pollutants from Long Range Transboundary Air Pollution*, World Health Organisation, Regional Office for Europe, Copenhagen, 2003.

14 U. G. Ahlborg, L. Lipworth and L. Titus-Ernstoff, et al., Organochlorine compounds in relation to breast cancer, endometrial cancer, and endometriosis: an assessment of the biological and epidemiological evidence, *Crit. Rev. Toxicol.*, 1995, **25**, 463–531.

15 P. O. Darnerud, S. Atuma and M. Aune, et al., Dietary intake estimations of organohalogen contaminants (dioxins, PCB, PBDE and chlorinated pesticides, *e.g.* DDT) based on Swedish market basket data, *Food Chem. Toxicol.*, 2006, **44**, 1597–1606.

16 J. E. McGraw and D. P. Waller, Fish ingestion and congener specific polychlorinated biphenyl and *p,p*-dichlorodiphenyldichloroethylene serum concentrations in a great lakes cohort of pregnant African American women, *Environ. Int.*, 2009, **35**, 557–565.

17 S. Tao, F. L. Xu and X. J. Wang, et al., Organochlorine pesticides in agricultural soil and vegetables from Tianjin, China, *Environ. Sci. Technol.*, 2005, **39**, 2494–2499.

18 S. Harrad, S. Hazrati and C. Ibarra, Concentrations of polychlorinated biphenyls in indoor air and polybrominated diphenyl ethers in indoor air and dust in Birmingham, United Kingdom: implications for human exposure, *Environ. Sci. Technol.*, 2006, **40**, 4633–4638.

19 K. Vorkamp, M. Thomsen and M. Frederiksen, et al., Polybrominated diphenyl ethers (PBDEs) in the indoor environment and associations with prenatal exposure, *Environ. Int.*, 2011, **37**, 1–10.

20 A. Burgess and H. L. Sweetman, The residual property of DDT as influenced by temperature and moisture, *J. Econ. Entomol.*, 1949, **42**, 420–423.

21 J. S. Colt, J. Lubin and D. Camann, et al., Comparison of pesticide levels in carpet dust and self-reported pest treatment practices in four US sites, *J. Expo. Anal. Environ. Epidemiol.*, 2004, **14**, 74–83.

22 J. S. Colt, R. K. Severson and J. Lubin, et al., Organochlorines in carpet dust and non-Hodgkin lymphoma, *Epidemiology*, 2005, **16**, 516–525.

23 M. Abb, J. V. Breuer and C. Zeitz, et al., Analysis of pesticides and PCBs in waste wood and house dust, *Chemosphere*, 2010, **81**, 488–493.

24 D. R. Fortune, F. D. Blanchard, W. D. Ellenson and R. G. Lewis, *Analysis of Aged Indoor Carpeting to Determine the Distribution of Pesticide Residues Between Dust, Carpet and Pad Compartments*, EPA 600/R-00/030 and EPA 600/R-00/030, 2000. Ref Type: Electronic Citation.

25 H. M. Hwang, E. K. Park and T. M. Young, et al., Occurrence of endocrine-disrupting chemicals in indoor dust, *Sci. Total Environ.*, 2008, **404**, 26–35.

26 Agency for Toxic Substances and Disease Registry, *Toxicological Profile for DDT, DDE and DDD*, US Department of Human Health Services, 2002, Ref Type: Report.

27 B. Brunekreef and S. T. Holgate, Air pollution and health, *Lancet*, 2002, **360**, 1233–1242.

28 R. A. Rudel, D. E. Camann and J. D. Spengler, et al., Phthalates, alkylphenols, pesticides, polybrominated diphenyl ethers, and other endocrine-disrupting compounds in indoor air and dust, *Environ. Sci. Technol.*, 2003, **37**, 4543–4553.

29 M. H. Ward, J. S. Colt and C. Metayer, et al., Residential exposure to polychlorinated biphenyls and organochlorine pesticides and risk of childhood leukemia, *Environ. Health Perspect.*, 2009, **117**, 1007–1013.

30 P. J. Landrigan, Risk assessment for children and other sensitive populations, *Ann. N. Y. Acad. Sci.*, 1999, **895**, 1–9.

31 *Highlights of the Child-Specific Exposure Factors Handbook (Final Report)*, US Environmental Protection Agency, Washington, DC, EPA/600/R-08/135, 2009, Ref Type: Report.

32 S. Harrad, C. Ibarra and M. Diamond, et al., Polybrominated diphenyl ethers in domestic indoor dust from Canada, New Zealand, United Kingdom and United States, *Environ. Int.*, 2008, **34**, 232–238.

