Trends in Inhalation Exposure to Hydrocarbons among Commercial Painters in the Netherlands

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Introduction

In 1995, ARBOUW (the Dutch national institute for occupational safety and health in the construction industry) has put in place a health surveillance programme designed to prospectively follow the health of commercial painters in the Netherlands. Part of the reason for establishing the program was the evidence that chronic toxic encephalopathy [Feldman et al., 1999; Bruhn et al., 1981; Arlien-Soborg et al., 1979], hearing loss [Sliwinska-Kowalska et al., 2001], decreased semen quality [Xiao et al., 1999], neuroblastoma in offspring [de Roos et al., 2001], neurological deficits [Dick et al., 2000], neurobehavioral symptoms [Chen et al., 2001; Triebig et al., 2000; Axelson et al., 1976], dementia [Mikkelsen, 1980] and decreased respiratory function (e.g. airflow obstruction) [Schwartz and Baker, 1988] among painters has been associated with exposure to hydrocarbons in previous studies. A case study of three painters with 25 to 46 years of exposure to solvents, presenting symptoms of Parkinson syndrome with pyromidal features, was described in the Netherlands in 1999 [Hageman et al., 1999]. Therefore, one of the goals of the health surveillance programme is to investigate the relationship between long-term exposure to organic solvents and chronic toxic encephalopathy. We set out to develop a database of exposure measurements that can be used as a tool for exposure assessment among commercial painters in the Netherlands. Results were reported in full elsewhere [Burstyn and Kromhout, 2002].

Methods

Building the exposure database

Seven studies were identified, which had sufficiently detailed information on airborne exposure levels among commercial painters for inclusion in the database. These were selected after consulting the peer-reviewed scientific literature and internal reports from leading research institutions in the field of occupational hygiene in the Netherlands. The data from one of the 1991 surveys were lost, leaving only six studies to be included in the database [Hokse and Sturm, 1995; Jongeneelen and Scheffers, 1982; Hilhorst et al., 2000; Pool and Veenstra, 1984; Veenstra et al., 1984; Boeckhout, 1999]. A database management system was created using a Microsoft Access 97 application. Contextual information was coded by investigators from the narrative descriptions of the workplaces where measurements were collected, which were supplied by the authors in the original reports. Only personal measurements were considered in the analysis.

Analysis of historical exposure trends

All concentrations that were reported to have been below the limit of detection were replaced with values equal to half of the concentrations cited in the original reports. Because we aimed to pool data across data sets, we selected agents that were measured in as many surveys and samples as possible as dependent variables. Correlation among the potential dependent variables was examined using both a Persons correlation and principal component analysis. Mixed-effects models were constructed to identify trends in exposure among all the exposure measurements gathered into the database. Potential determinants of exposure were treated as fixed effects. Worker was treated as a random effect, since there were multiple measurements from individual painters in some surveys. Analyses were carried out in SAS 6.12 (SAS Institute, Cary, NC, USA).

Results

Historical trends in exposure

304 solvent exposure measurements were collected between 1980 and 1999, providing data from 137 workers. Measurements of more than 4000 concentrations of individual chemicals were collected. Most measurements were collected during application of solvent-based paints, using randomised sampling strategies (Table 1). Measurements during application of solvent-based, but not water-based, paints were available for all time periods, enabling estimation of time trends in exposure due to use of solvent-based paints only. Data were available for painting done in houses and in shipyards, during spray-painting and manual painting with a brush and/or a roller.

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TABLES

Table 1 Description of data sets collected into a database of historical exposures among commercial painters in the Netherlands

Year	Number of samples collected during use of different paints: All	SB *	H2 †	WB‡	Spray-painting **	Workers	Sampling days	sampling strategy: Random day	Random worker	Other features	Location	Average sampling duration (minutes)
1980	45	39	5††	1	5	45	12	Yes	Yes	Random	House	Full shift
1982-83	22	22	0	0	0	21	20	No	No	worst case	House	221
1984	4	4	0	0	2	2	2	Yes	No	task-based	Shipyard	168
1994	95	47	0	42	0	33	19	Yes	Yes	avoid consecutive days; all workers available	House	215
1998	10	10	0	0	0	6	3	Yes	Yes	random	House	176
1999	128	25	44	6	24	30	53	Yes	Yes	4 measurement periods/year	Shipyard	461

** solvent-based: alkyd-based, synthetic wall paints, traditional turpentine paints, chloro-rubber and unspecified solvent-based paints.*
† high-solid or two-component
‡ water-based (acrylate)
*** number of samples collected during spray-painting (versus manual painting with roller and/or brush)*
†† two-component paint only, since high-solid paint was only introduced in 1990's

Table 2 Exposure levels during use of different paints

Paint type	Hydrocarbon	N*	AM† (mg/m³)	GM‡ (mg/m³)	GSD**	Min† (mg/m³)	Max‡‡ (mg/m³)
All	Toluene	304	1.06	0.09	11.47	0.004	43.00
	Xylene †††	259	11.47	1.55	9.37	0.005	391.72
	Ethyl-Benzene	259	2.87	0.30	11.94	0.002	86.00
	n-Hexane	223	0.10	0.01	4.06	0.007	10.93
	n-Decane	245	5.85	0.43	13.87	0.005	210.00
Water-based (acrylate)	Toluene	49	0.61	0.12	7.24	0.004	9.43
	Xylene †††	48	1.15	0.30	6.17	0.011	13.13
	Ethyl-Benzene	48	0.23	0.05	8.67	0.002	1.73
	n-Hexane	48	0.04	0.03	2.22	0.007	0.33
	n-Decane	48	1.75	0.57	5.42	0.010	23.46
High-solid and (n=11) two-component (n=38)	Toluene	49	0.47	0.05	9.78	0.004	4.00
	Xylene †††	44	25.99	5.64	6.49	0.094	391.72
	Ethyl-Benzene	44	3.86	1.02	6.05	0.021	42.32
	n-Hexane	44	0.08	0.01	4.85	0.007	1.62
	n-Decane	44	0.50	0.13	5.99	0.005	4.80
Other solvent-based ***	Toluene	147	1.93	0.24	8.94	0.004	43.00
	Xylene †††	108	15.26	4.31	5.16	0.085	233.50
	Ethyl-Benzene	108	4.95	1.04	6.36	0.023	86.00
	n-Hexane	72	0.20	0.02	4.26	0.007	10.94
	n-Decane	94	13.90	2.69	8.76	0.006	210.00

* sample size
† arithmetic mean
‡ geometric mean
** geometric standard deviation
†† minimum value
‡‡ maximum value
*** alkyd-based (n=116), synthetic wall paints (n=8), traditional turpentine paints (n=4), chloro-rubber (n=16) and unspecified solvent-based (n=3) paints
††† the sum of othro-, para- and meta-xylene

Toluene, xylene, ethyl-benzene, n-decane and n-hexane were measured most commonly among Dutch commercial painters, and were consequently used as a marker for the solvent exposure of these workers. These levels are summarised in Table 2. Concentrations of these hydrocarbons followed frequency distributions well approximated by log-normal distributions, and thus all subsequent analyses were performed with log-transformed concentrations. Xylene and ethyl-benzene concentrations tended to be higher during application of solvent-based paints. Patterns of exposure levels for toluene, n-hexane and n-decane did not indicate clear differences between those for water-based and solvent-based paints. The Dutch occupational exposure limit for toluene (150 mg/m^3) was not exceeded in any of the measurements. However, the xylene exposure limit of 210 mg/m^3 was exceeded in two measurements, which were collected during painting with solvent-based paints.

Examination of correlation among selected hydrocarbon concentrations in the air (data not shown) revealed that only xylene and ethyl-benzene were highly correlated (r=0.97, p<0.0001, n=233). However, there was a weak to moderate positive correlation among pairs of all five hydrocarbons (toluene, xylene, ethyl-benzene, n-decane and n-hexane). Principal component analysis revealed that two sources of variability accounted for 79% of multiple correlation among the selected hydrocarbons. The first principal component was approximately equally positively associated, as judged by the eigenvector, with all five hydrocarbons. The second principal component was primarily positively associated with n-hexane, and negatively associated with xylene and ethyl-benzene.

On the basis of these considerations, and taking into account the distribution of individual hydrocarbon concentrations, we selected toluene as a marker of solvent exposure in modelling historical exposure trends (Table 3). Between 1980 and 1999 toluene exposures measured during application of solvent-based paints have declined, on average, 12% per year, resulting in the halving of exposure levels approximately every 5.5 years. Solvent-based paints were associated with an approximately 17 times greater toluene exposure relative to those circumstances where paints were not used (e.g. clean-up or preparation of surfaces for painting). Water-based paints were associated with an approximately doubled toluene exposure relative to the circumstances were no paints were used. As could have been expected, samples collected in small rooms (<50 m^3, also identified as worst-case sampling in one survey) were associated with a near-doubling of toluene concentration, compared to samples collected in

Table 3 Determinants of exposure to toluene (log(mg/m3), N=304, k=137*) among commercial painters: results of mixed effects models with worker as random effect and compound symmetry covariance structure.*

Determinant of exposure	Range or count ‡‡	β†	SE‡	p(β≠0)**
Intercept	--	-2.84††	0.36	0.0001
Year (since 1980) Solvent-based paint	0-19	-0.12	0.02	0.0001
Solvent-based paint (1) Vs. no paint (0)	196	2.82	0.44	0.0001
Water-based paint (1) Vs. no paint (0)	49	0.74	0.41	0.07
Small room (1) Vs. large room or Outdoors (0)	100	0.45	0.25	0.07
Shipyard (1) Vs. house painting (0)	132	-2.25	0.36	0.0001
Spray-painting (1) Vs. manual (0)	31	1.68	0.39	0.0001
Covariance components		Estimate (% explained by model***)	SE‡	p†††
Between worker		0.49 (86)	0.18	0.0070
Within worker (day-to-day)		2.22 (11)	0.22	0.0001

* *N = number of measurements; k = number of workers; † regression coefficient for fixed effect; ‡ standard error of β or estimate of covariance component; ** t-test for the fixed effect; †† exposures when paint was not used (n=59 for toluene exposure), during manual house painting under representative conditions; ‡‡ for determinants of exposure, counts for dummy variables (number of "yes") and ranges for continuous variables; *** percent of covariance component explained, relative to model with only random effect; ††† p-value of Wald test for covariance component being equal to zero*

large rooms and outdoors. House painting was associated with a 9-fold increase in toluene concentration relative to shipyard painting (assuming similar type of application). Spray-painting was associated with a factor of five increase in toluene exposures relative to manual painting. Presence of local exhaust ventilation was not retained in the final model, because of confounding: it was associated with higher exposure concentrations, while negating the effect of small room volume. The toluene exposure model explained most of the between-worker variance (86%), reducing the residual range within which 95% of average exposure concentrations for individual workers are likely to fall ($_{BW}R_{0.95}$ [Rappaport, 1991]) from 1580 to 16. However, substantial day-to-day (within-worker) variance remained unexplained by our model. Both between- and within-worker variance components estimated in the mixed effects model were significantly different from zero. The trends in exposure to toluene during the predominant use of solvent-based paints in large room or outdoors predicted by our models are illustrated in Figure 1.

surveillance program have recently been highlighted [Fransman et al., 2002]. Quantitative exposure estimates that recognise time trends in exposure are needed to facilitate exposure-response modelling and accurate risk assessment through longitudinal studies.

Toluene appears to be a promising proxy of aromatic hydrocarbon solvent exposure, since it appeared to correlate well with other aromatic hydrocarbons. However, our data was limited in its ability to explore the relation between aromatic and aliphatic hydrocarbons, since aliphatic hydrocarbons (such as n-hexane) were not monitored in all production circumstances where toluene was determined. Thus, it is recommended that the relationship between aliphatic and aromatic hydrocarbon exposure during painting should be explored in future studies. This is especially important, since we did identify a significant second principal component among hydrocarbon exposures, indicating that relative composition of hydrocarbon emissions can vary substantially among different production circumstances.

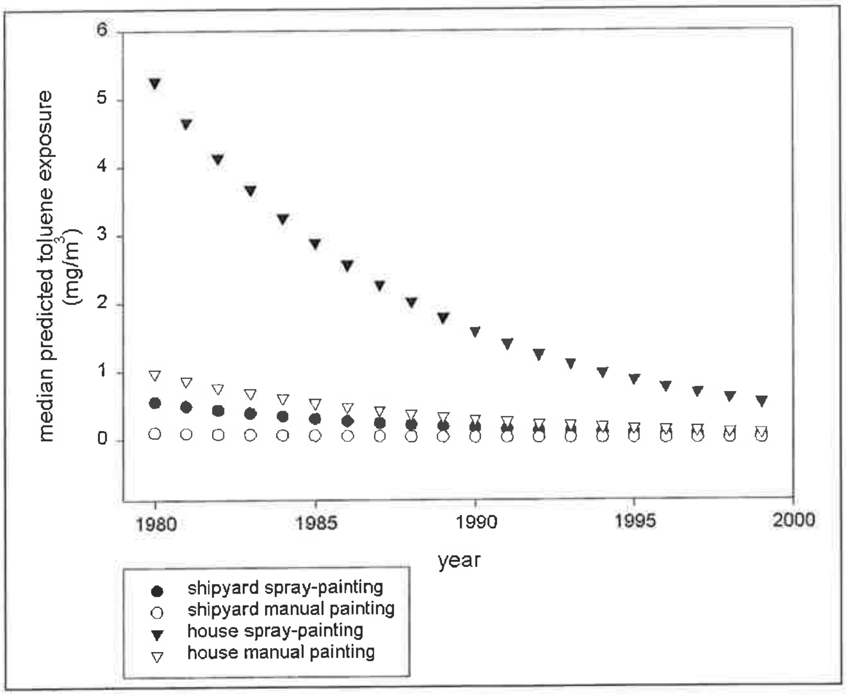

Figure 1 Predicted trends in exposure to toluene of commercial painters predominantly applying of solvent-based paints either in large rooms (>50 m³) or outdoors.

We observed that exposure levels of painters using solvent-based paints have declined by a factor of 11 over a 20-year period in the Netherlands. This reflects the decline in solvent content of solvent-based paints and the introduction of water-based paints that have become more widely used since the 1990's. A historical decline in exposures to solvents among painters has been reported recently, but not quantified [Caldwell et al., 2000].

In any exposure assessment that applies our results, the modelled exposure intensity estimates would have to be adjusted for use of respiratory protective equipment. To achieve this, we propose to use previously estimated protection factors for painters exposed to mixed solvents: zero for dust masks, 0.65 for cartridge and 0.90 for supplied air respirators [Fidler et al., 1987].

Discussion

Our study is the first attempt to use historical exposure measurements to model determinants of exposure among commercial painters, and is the first report of an exposure database specifically developed to support a health surveillance programme among commercial painters. Previous attempts to model long-term exposures to mixed solvents among painters either relied on an expert assessment approach, or simply used duration of exposure as a surrogate of dose [Fidler et al., 1987]. Problems with application of expert assessment of solvent exposure in an ongoing health

The toluene exposure model has important limitations. First, categorical variables identified as determinants of exposure do not reflect the complexity of the workplaces. Second, the estimation of the effect of local exhaust ventilation was confounded by its being used when exposures were expected to be high (such as in confined spaces in the hull of ships). Third, we did not observe differences between exposure levels during painting in large rooms and outdoors. This may reflect the fact that, at present, the use of solvent-based paint is permitted in outdoors application, but not in indoor application in the Netherlands.

In conclusion, using toluene as a proxy for total solvent exposure, we developed an exposure model that can be used to predict the intensity of inhalation exposure to aromatic solvents for the years 1980-1999 in a health surveillance programme among commercial painters in the Netherlands. The lessons learned from this project have important implications for creation of similar databases for either retrospective or prospective occupational exposure data collection, such as the proposed National Occupational Exposure Database project in the Netherlands.

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