

# Trends in Occupational Exposure to Styrene in the European Glass Fibre-Reinforced Plastics Industry

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**Aim:** This study presents temporal trends of styrene exposure for workers in the European glass fibre-reinforced plastics (GRP) industry during the period 1966–2002.

**Methods:** Data of personal styrene exposure measurements were retrieved from reports, databases and peer-reviewed papers. Only sources with descriptive statistics of personal measurements were accepted. The styrene exposure data cover personal air samples and biological monitoring data, that is, urinary styrene metabolites (mandelic acid and/or phenylglyoxylic acid) and styrene in blood. Means of series of measurements were categorized by year, country, production process, job and sampling strategy. Linear mixed models were used to identify temporal trends and factors affecting exposure levels.

**Results:** Personal exposure measurements were available from 60 reports providing data on 24145 1–8-h time-weighted average shift personal air samples. Available data of biological exposure indicators included measurements of mandelic acid in post-shift urine (6361 urine samples being analysed). Trend analyses of the available styrene exposure data showed that the average styrene concentration in the breathing zone of open-mould workers in the European GRP industry has decreased on average by 5.3% per year during the period 1966–1990 and by only 0.4% annually in the period after 1990. The highest exposures were measured in Southern Europe and the lowest exposures in Northern Europe with Central Europe in between. Biological indicators of styrene (mandelic acid in post-shift urine) showed a somewhat steeper decline (8.9%), most likely because urine samples were collected in companies that showed a stronger decrease of styrene exposure in air than GRP companies where no biological measurements were carried out.

**Keywords:** air monitoring; biological monitoring; European GRP industry; linear mixed model analysis; mandelic acid (MA); occupational exposure; phenylglyoxylic acid (PGA); Styrene (CAS 100-42-5); temporal trends

## INTRODUCTION

The evaporation of styrene from unsaturated polyester resin into the work environment during processing in the Glass fibre-Reinforced Plastics (GRP) can result in significant exposures to styrene. Various publications and reports describe possible adverse health effects among workers within the industry. In the USA, the American Conference of Governmental Industrial Hygienists (ACGIH) considers the most critical adverse effects of styrene are irritation

and effects on the central and peripheral nervous system. The ACGIH has proposed that a threshold limit value of 20 ppm [ $\approx 87 \text{ mg m}^{-3}$ ; 8-h time-weighted average (TWA)] and 40 ppm ( $\approx 174 \text{ mg m}^{-3}$ ; short-term exposure limit) in the workplace will minimize these effects (ACGIH, 2001). In Europe, 8-h time-weighted occupational exposure limits for styrene vary between 20 ppm (Germany) and 100 ppm (UK). Short-term occupational exposure limits range from 75 (Sweden) to 250 ppm (UK). On behalf of the European Union (EU), the Health and Safety Executive (HSE) in the UK is preparing an extensive risk assessment of styrene including a review of the available studies on many toxicological end points.

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This document contains an overview of the actual occupational exposure limits for styrene in European countries (HSE, 2007).

Several techniques and product developments have been introduced to reduce styrene emissions in the GRP industry. In addition, both technical and organizational control measures have been implemented in many companies across Europe. Introduction of control measures has been supported by provision of information on the properties of unsaturated polyester resins, available processing techniques and technical control measures (Aurer and Kasper, 2003). While a recent publication by the German Institute for workers protection (BGIA, 2006) provides an overview of state-of-the-art technical protective measures relating to the handling of styrene. Some examples of these product developments and control measures are:

- Introduction of resins with lower styrene content. A typical resin contains ~40% styrene. Dicyclopentadiene (DCPD)-based resins contain ~35% styrene. Recent developments have been published in which resins are described with contents of 25–30% by weight.
- The use of film-forming additives in the resin to limit the evaporation of styrene. Resins that contain a film-forming additive, such as paraffin, are called low-styrene emission (LSE) resins.
- The use of so-called light curable resins. This type of resins quickly forms a film of cured resin on top that will stop styrene emission almost instantly. The use of this type of resin is restricted to transparent products only.
- Introduction of closed-mould techniques, such as vacuum injection and so-called resin transfer moulding.
- Controlled spraying minimizing over-spray.
- Extended use of personal protection equipment like clean-air helmets, reusable respirators, gloves, etc.

To assess the risk of exposure to styrene and to enable critical evaluations of former publications on styrene health effects, it is essential to have accurate exposure estimates of workers over a long time period in the European GRP industry.

While over the last three decades numerous reports and publications have appeared describing measurements of exposure to styrene, it has proven difficult to develop a good understanding of styrene exposures in the GRP industry over this period. These difficulties include differences in sampling strategy, sampling and analytical methods, sampling time, sampling year, geographical areas, job category and type of production process.

In this study, manuscripts and publications reporting data on styrene exposure in the European GRP industry have been collected to produce exposure data

and information on production process characteristics, country, year, job category and sampling strategy. This information was collected to analyse temporal trends in occupational styrene exposure and to estimate exposure of workers in various jobs in open- and closed-mould production of GRP in Europe during a more than 30-year period from 1966 to 2002.

## METHODS

### *Data collection*

Sources of workers' exposure to styrene in the European GRP industry since 1970 were scientific peer-reviewed articles and various reports. Peer-reviewed articles were searched through the literature databases MEDLINE and TOXNET. In addition, company reports with exposure data were available from conseil européen des fédérations de l'industrie chimique (CEFIC)/Styrene Steering Committee. Statistical descriptives of styrene exposure data from databases in Europe, for example, NEDB-UK, MEGA-Germany (update till 2002), CEFIC (six European countries), Norway, Denmark, Finland and France, as far as they were published, were included in this review.

A first screening of available literature and reports resulted in a selection of 94 publications with information on styrene exposure from European GRP workers (see Appendix 1). After studying these 94 articles and reports in detail, 34 were put aside as they did not contain information on descriptive statistical parameters (e.g. arithmetic or geometric means or median and (geometric) standard deviation or percentiles or ranges) or contained data that were already available from other publications.

Two experienced industrial hygienists carefully screened each of the remaining 60 articles and reports for information on styrene exposure measurements. All this information was incorporated in an MS Access database elaborated for the occasion. The variables included in the styrene exposure database are presented in Table 1.

All of the retrieved exposure data were converted to the same units of measurement: air monitoring data:  $\text{mg m}^{-3}$  and urinary metabolites:  $\text{mg g}^{-1}$  creatinine.

The arithmetic mean (AM) was used for the comparison of exposure data from various sources and to analyse temporal trends. When the AM was not reported in the original article or report, we calculated the AM from the available descriptive statistics assuming a lognormal distribution of the exposure data. The majority of the publications and reports only contained descriptive statistics on styrene exposure. Results of individual measurements from which mean values of styrene exposure were calculated were not available.

Table 1. Variables included in the styrene exposure database

1	Region/country	12 classes: EU countries.
2	Type of production process	Four classes: open mould, closed mould, other, not reported.
3	Product	Five classes: boats, small products, medium products, not specified, not reported.
4	Type of resin	Three classes: DCPE, LSE, not reported.
5	Purpose of study	Six classes: research, evaluation controls, testing compliance company, testing compliance regulator, concern, not reported.
6	Ventilation	Three classes: mechanical room ventilation, local ventilation, not reported.
7	Personal protective equipment	Five classes: respiratory, gloves, both, none, not reported.
8	Job category	23 classes, see Table 2.
9	Year of sampling	35 years: 1966–2002.
10	Sampling strategy	Three classes: random, worst case, not reported.
11	Exposure indicator	Three classes: air samples, biological samples, both
12	Type of air sampling	Four classes: personal, area, source oriented, not reported.
13	Air sampling method	Four classes: passive, active, real time, not reported.
14	Air sampling duration	Continuous, hours
15	Concentration styrene in air	Continuous, descriptive statistics of series.
16	Sampling time biological sample	Five classes: post-shift, during shift, next day, other, not reported.
17	Metabolite in urine	Three classes: MA, PGA, MA + PGA.
18	Concentration metabolite in urine	Continuous, descriptive statistics of series.
19	Concentration styrene in blood	Continuous, descriptive statistics of series.

### Statistical analyses

A statistical analysis, applying linear mixed effect models, was carried out using the mean values of styrene in breathing zone (1–8 h samples) and mandelic acid in post-shift urine samples of open-mould workers. As dependent variables the log-transformed value of the ‘mean air concentration’ and the ‘mean mandelic acid in post-shift urine’ were used. The effect of ‘year since 1966’ was analysed while taking into account the job category, the region in Europe and potential confounding factors like ‘measurement purpose’, ‘sampling strategy’ and ‘sampling method’.

Visual inspection of the exposure data (see Fig. 1) indicated that pre-1990 the exposure decline might have been at a higher rate than post-1990. This was confirmed in a formal analysis with natural splines. This analysis indicated that a piecewise linear model, sometimes called a ‘broken-stick’ model, would fit the exposure data very well. Therefore, we included an additional determinant ‘year since 1990’ to the model to allow for a different temporal trend after 1990. Grouping of EU countries in regions was as follows—Region North: Norway, Sweden, Finland and Denmark; Region Central: UK + Ireland, Benelux, Germany and Switzerland; Region East: countries classified as ‘Eastern countries’, e.g. Poland and Hungary; Region South: France, Italy, Spain and Portugal.

Given the fact that analyses were performed on aggregated data, a weighted analysis was completed with the number of observations underlying each mean air concentration as weighting unit. The majority of the studies lacked information on exposure variability and therefore weighing for observed variability in the individual measurement series was

no option. Given the potential dependency of multiple observations from the same survey, ‘survey’ was used as a random effect in the linear mixed effects models.

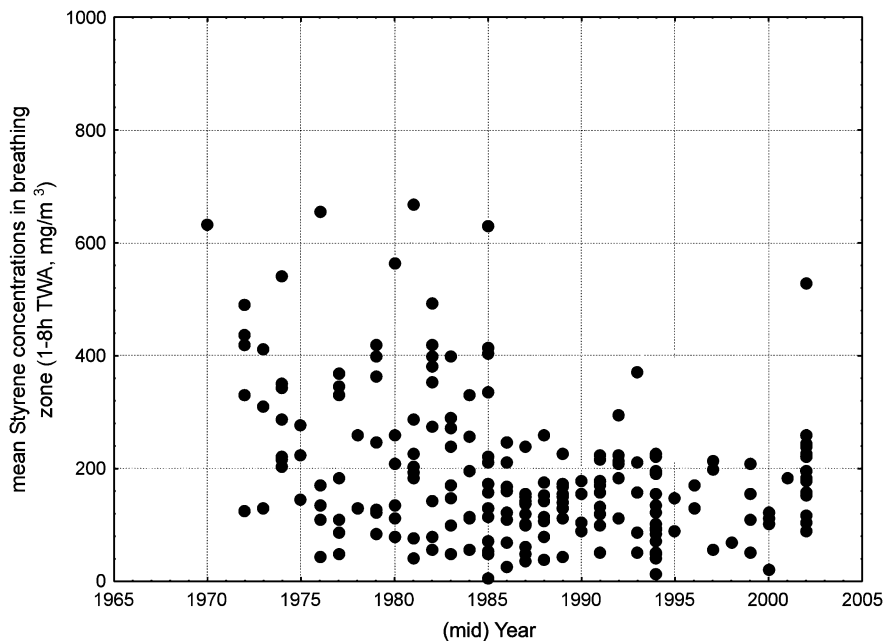
Model building started with an unconditional model (so-called naive model) with only the random effect of survey. Next, main exposure-determining factors like ‘year since 1966’, ‘year since 1990’, ‘job category’ and ‘region’ were step-by-step added to the model. Interaction effects between ‘year since 1966’ or ‘year since 1990’ and factors like ‘region’ and ‘job category’ were considered after this. Finally, the effect of potential confounding factors like measurement method, sampling strategy, etc. described earlier was modelled. Only factors that reached a significance level  $<0.05$  and/or affected the coefficients of other factors by  $>10\%$  were kept in the multivariate model.

The explained variability of the model was estimated by comparing the within- and between-survey variability of the naive model (with only survey as a random factor) with their estimates in the full mixed model with the fixed determinants and the random survey factor. This is a common approach in linear mixed models that was introduced by Burstyn et al. (2000). Analyses were carried out with SAS 9.1 software (SAS Institute, Cary, NC, USA).

## RESULTS

### Retrieved data on styrene exposure

Concentrations of styrene in breathing air were classified by sampling time in two classes: short-term



**Fig. 1.** Average breathing zone styrene concentrations of European GRP workers applying open-mould techniques from 1966 to 2002 (each point represents the AM of a measurement series;  $N = 213$ ;  $1 \text{ mg m}^{-3} \approx 0.23 \text{ ppm}$ ).

samples ( $<1 \text{ h TWA}$ ) and shift samples ( $=1\text{--}8 \text{ h TWA}$ ). The number of short-term samples (data not shown) was limited in comparison to shift samples: 12 and 268 mean values, respectively. These mean values were based on respectively 3028 short-term samples and 24145 shift samples. Further statistical analysis of the data was restricted to 1–8 h TWA shift samples only. The majority (70%) were estimated to have lasted 8 h.

An overview of the retrieved air samples (1–8 h TWA) and biological exposure indicator (mandelic acid in post shift urine) in the period 1966–2002 is presented in Table 2. In this table, the estimated use of polyester resin in Europe in 2002 is also presented. Data on polyester resin use were retrieved from unpublished files from CEFIC. Data on use of polyester resin in the early '70s were not available.

Only a few of the 94 publications and reports that we screened contained information on use of personal protective equipment (PPE) and ventilation circumstances during the measurements.

The sampling methods were either not reported (56%) of the passive diffusion type (26%) or with an active pump-based method (16%). The actual purpose of the surveys differed a lot with 33% for research health-related purposes, 27% because of raised concern and 11% for compliance reasons. For more than a quarter (26%) of the surveys the reason was not reported. The actual measurement strategy was only seldom known. Random sampling was applied in 29% of the surveys; worst-case sampling only took place in 9% of the surveys. For 62% of

the survey, no actual information on the measurement strategy was found.

#### *Data by country in EU*

Most of the retrieved air sampling data were from the Nordic countries (37%), Germany (24%), France (15%) and UK (13%). Data on styrene in air samples from workers in Spain, Portugal and Eastern Europe were rare ( $<2\%$ ), while in Italy, biological monitoring was particularly used to assess occupational exposure to styrene. Comparison of the retrieved exposure data with the use of polyester resin in the various countries shows that relatively many air sampling data were collected in the Nordic countries (37% versus 7% of polyester resin use). Styrene exposure data from Spain and Portugal were very limited although these countries used  $\sim 16\%$  of the polyester resin in 2002 (see Table 2).

#### *Data by production technique and job category*

More than 90% of the retrieved data on styrene exposure in the period 1966–2002 were obtained from companies using an open-mould production process, mainly the laminating/spray-up technique (see Table 2). Data on styrene exposure in closed-mould production facilities of GRP products were available for only 2% of all shift samples. It is noted that in 2002, open-mould production techniques accounted for  $\sim 53\%$  of the total European resins use with  $\sim 40\%$  of the total amount of polyester resin used in closed-mould techniques (see Table 2).

Table 2. Overview of retrieved exposure data by (a) country, (b) production technique and (c) job category in the period 1966–2002

	Use of polyester resin (in 2002)		Air samples (shift: 1–8 h)			Urine samples (mandelic acid—post-shift)		
	Tons	%	Number of retrieved AMs	Number of samples		Number of retrieved AMs	Number of samples	
<b>(a) By country/region</b>								
Norway	37.800	7	16	5023	21%	0	0	0%
Sweden			44	1900	8%	2	23	0.4%
Finland			26	979	4%	10	1220	19%
Denmark			7	886	4%	4	155	2.4%
UK + Ireland	85.833	16	26	3237	13%	3	53	0.8%
Benelux	22.200	4	17	344	1%	3	52	0.8%
Germany	85.700	16	17	5761	24%	5	84	1%
France	82.100	15	10	3611	15%	2	284	4%
Switzerland	5.300	1	1	90	0.4%	0	0	0%
Eastern Europe	?	?	16	278	1%	3	28	0.4%
Spain + Portugal	85.033	16	2	47	0.2%	3	42	0.7%
Italy	116.000	22	67	1808	8%	8	4420	69%
Various countries	—	—	19	211	0.9%	0	0	0%
<b>Total</b>	530.600 <sup>a</sup>	100	268	24145	100%	43	6361	100%
<b>(b) By production technique</b>								
Open mould	280.400	53	213	22718	94%	37	4957	78%
Closed mould <sup>b</sup>	210.700	40	19	568	2%	0	0	0%
Other technique <sup>c</sup>	39.500	7	28	705	3%	1	762	12%
Not reported	—	—	8	154	1%	5	642	10%
<b>Total</b>	530.600~	100	268	24145	100%	43	6361	100%
<b>(c) By job category (specified for open mould only)</b>								
1.1. Laminating—lay up			31	2951	13%	7	2513	51%
1.2. Laminating—spray up			30	845	4%	6	585	12%
1.3. Laminating—non-specified			69	11 269	50%	11	206	4%
1.4. Gel or top coating			6	292	1%	0	0	0%
1.5. Rollers			0	0	0%	1	63	1%
1.6. Filament winding			12	231	1%	1	33	1%
1.7. Repair putties—use			2	194	1%	0	0	0%
1.8. Variable open mould			58	6563	29%	11	1557	31%
1.9. Variable open mould—construction industry			2	291	1%	0	0	0%
1.10. Not specified			3	82	0.4%	0	0	0%
<b>Total</b>			213	22718	100%	37	4957	100%

<sup>a</sup>Total use of resin in 2002 includes the usage in Greece (3933 tons) and Austria (6700 tons).

<sup>b</sup>Seven job categories: laminating continuous, injection techniques, pultrusion, cold press, SMC and BMC—moulding (hot press), relining—*in situ*, variable closed mould.

<sup>c</sup>Five job categories: finishing activities, SMC and BMC production, polymer concrete production, repair putties production, other.

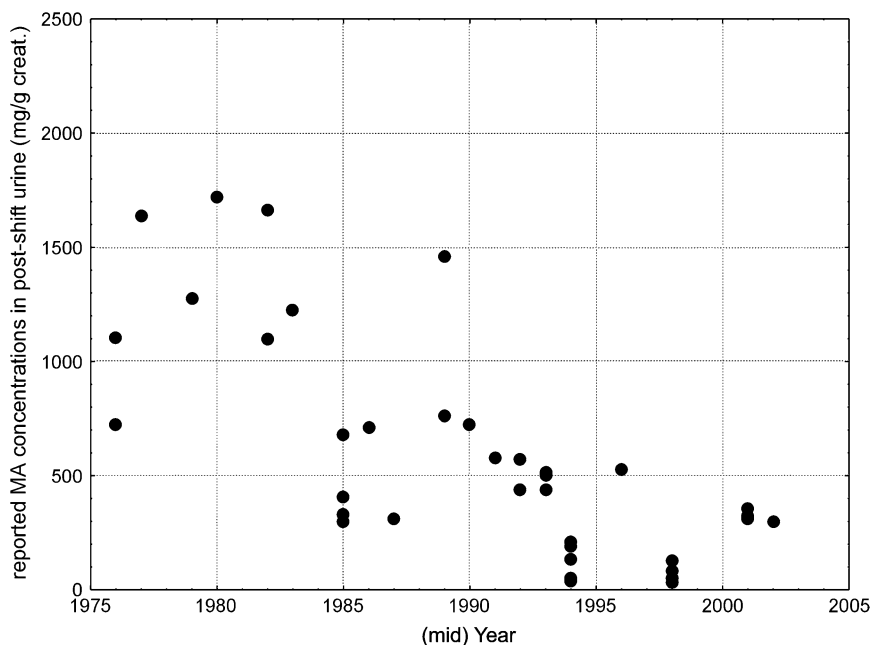
### Styrene exposure in the open-mould industry

Figure 1 shows the retrieved mean styrene concentrations in breathing zone of open-mould workers from all over Europe, from 1966 onwards for each individual survey. These mean values ( $n = 213$ ) are based on 22718 individual shift samples (1–8 h TWA). The figure shows that reported mean styrene concentrations in '70s and early '80s ranged up to ~650 mg m<sup>-3</sup> (150 ppm). In the '90s, the average styrene exposures of open-mould workers in Europe

tend to be lower: 50–250 mg m<sup>-3</sup> (12–58 ppm). Figure 1 also shows that the number of retrieved mean values per 5-year period is rather constant.

### Styrene exposure in the closed-mould industry

Data on the styrene exposure of European closed-mould workers are limited. We only retrieved 19 mean values of styrene concentrations (1–8 h TWA) in the breathing zone, based on 568 samples (data are not shown). These data indicate that styrene exposure in



**Fig. 2.** Average mandelic acid concentrations in post-shift urine of European GRP workers applying open-mould techniques from 1976 to 2002 (each point represents the AM of a measurement series;  $N = 37$ ).

closed-mould workers were substantially lower than in open-mould workers. In the '90s, the mean styrene concentration in the working atmosphere of closed-mould workers ranged between 10 and 90  $\text{mg m}^{-3}$  (2–21 ppm).

#### *Urinary indicator of exposure to styrene*

More than 95% of the retrieved biological monitoring data are based on measurement of the styrene metabolites, mandelic acid and/or phenylglyoxylic acid, in urine. The most popular urinary marker is mandelic acid in post-shift urine. We retrieved 43 mean values based on 6361 urine samples. As indicated in Table 2, the majority of biological monitoring samples, ~80%, came from companies applying open-mould techniques. The AM values of mandelic acid in post-shift urine of European workers applying open-mould techniques (37 mean values based on 4957 urine samples) from 1966 onwards are shown in Fig. 2.

#### *Statistical modelling of exposure data*

A temporal trend analysis of styrene exposure was carried out using the measurements from workers in the open-mould production process. In total, 213 mean styrene concentrations in the breathing zone (1–8 h TWA) and 37 mean mandelic acid concentrations in post-shift urine were available for the modelling of the time trend. The distribution of these data over the years of sampling, regions and job categories is shown in Figs 1 and 2 and Table 2a,c.

The statistical analyses showed that 'year since 1966' and 'year since 1990', as well as 'region'

and 'job category' appeared to be strong predictors of mean exposure to styrene among open-mould process operators. Together, these four variables explained 57% of total variability in (weighted) mean styrene concentrations in the breathing zone of these European GRP workers. The interaction between 'year since 1966' or 'year since 1990' and 'region' was not statistically significant ( $P > 0.05$ ), indicating there were no significant differences in time trend between regions. Potential confounding factors like measurement strategy, purpose and sampling method appeared to have no significant influence.

The model implies a significant decline of styrene concentrations in the breathing zone of European GRP workers of 5.3% per year (calculation:  $100(1 - \exp(-0.053)) = 5.3\%$ ) during the period 1966–1990 ( $P < 0.0001$ ;  $n = 213$ ). After 1990, the decline is no longer apparent: 0.4% (calculation:  $100(1 - \exp(-0.053 + 0.049)) = 0.4\%$ ) (Table 3).

Regional differences in styrene exposure are shown in Fig. 3a. In this figure, the time trend of the styrene exposure of all laminators combined (job category 1.1, 1.2 and 1.3) is presented for each of the four European regions. This figure shows that the highest exposures were measured in the southern region and the lowest exposures in the northern part with central Europe somewhere in between. The available data indicate that the exposure was also relatively low in the eastern part of Europe. However, it is noted that this finding is based on only 12 surveys that were conducted in a relatively short period of 9 years (1989–1997).

Table 3. Exposure affecting factors for exposure to styrene among workers in open-mould production in the European GRP industry: results of linear mixed model with mean air concentrations weighted by number of observations as dependent variable ( $n = 213$ )

Variable	$\beta$ (Standard error)	$P$
<b>Intercept</b>	<b>6.08 (0.37)</b>	<b>&lt;0.0001</b>
Job category 1.1. Laminating—lay up	0.61 (0.35)	0.08
Job category 1.2. Laminating—spray up	0.45 (0.36)	0.21
<b>Job category 1.3. Laminating—non-specified</b>	<b>0.85 (0.34)</b>	<b>0.01</b>
Job category 1.4. Gel or top coating	0.29 (0.38)	0.46
Job category 1.6. Filament winding	-0.22 (0.40)	0.58
Job category 1.7. Repair putties—use	-0.63 (0.41)	0.12
Job category 1.8. Variable open mould	0.34 (0.34)	0.32
Job category 1.9. Variable open mould— Construction industry	0.61 (0.39)	0.12
Job category 4. Not reported	0	
<b>Region central</b>	<b>-0.28 (0.08)</b>	<b>0.00004</b>
<b>Region eastern</b>	<b>-0.79 (0.22)</b>	<b>0.00003</b>
<b>Region nordic</b>	<b>-0.74 (0.09)</b>	<b>&lt;0.0001</b>
Region southern	0	
<b>Year since 1966</b>	<b>-0.053 (0.007)</b>	<b>&lt;0.0001</b>
<b>Year since 1990</b>	<b>0.049 (0.012)</b>	<b>0.0001</b>
	$S^2 (S_{naive}^2)$	
Between-survey variance	0.00 (0.00) <sup>a</sup>	
Within-survey variance	9.39 (21.87) <sup>a</sup>	

<sup>a</sup>Estimates of variance components for naive model (only random survey effect). variables with  $P$ -level  $\leq 0.01$ .

Job category was also found to be a statistically significant and strong predictor of exposure to styrene in European GRP workers applying open-mould techniques (see Table 3). Figure 3b illustrates the differences in styrene exposures for five job categories in open-mould production. The highest exposed workers were those classified as 'laminating non-specified' (job category 1.3). The lowest exposed group was the group of filament winders (job category 1.6).

Estimated styrene concentrations in open-mould workers in the European GRP industry in the year 2003 based on the model presented in Table 3 can be found in Table 4.

#### *Mandelic acid in post-shift urine samples of open-mould workers*

Modelling the available data on mandelic acid in post-shift urine samples of open-mould workers shows that only 'year since 1966' and 'job category' are strong predictors of the mandelic acid in post-shift urine samples of European open-mould work-

ers. Together, these two variables explained 66% of the total variability in (weighted) mean mandelic acid concentrations in urine of these workers. 'Year since 1990', 'region' and potential confounding factors like 'measurement strategy' and 'purpose' appeared to have no influence ( $P > 0.05$ ).

To illustrate both the trend and differences between the job categories, the urinary mandelic acid concentrations of four job categories by year have been plotted in Fig. 4. The workers with the highest mandelic acid concentrations are those classified as 'laminating non-specified' (job category 1.3) and 'laminating spray-up' (job category 1.2). The lowest mandelic acid concentrations are found in the 'variable open-mould' workers (job category 1.8).

The estimated annual decline calculated with job category and year as fixed effects was 8.9% in the period 1976–2002 ( $P < 0.0001$ ;  $n = 37$ ). This decline is substantially higher than the annual decline found for styrene concentrations in the breathing zone of open-mould workers (pre-1990: 5.3%, post-1990: 0.4%;  $n = 213$ ). Whether this steeper decline is caused by, for example, a more frequent use of PPE cannot be discerned from the data because information on the use of PPE by monitored workers was not available.

Of the 37 surveys with measurements of mandelic acid in post-shift urine, 24 surveys also have combined measurements of styrene concentrations in breathing zone air. Additional analyses of this sub-set of 24 surveys showed during the period 1977–1996 an annual decline of 7.1% ( $P < 0.05$ ) in post-shift urinary levels of mandelic acid and 7.7% ( $P < 0.01$ ) decline in styrene air concentrations ( $P = 0.005$ ;  $n = 24$ ). Apparently, urine samples have been collected in companies with a steeper decline of styrene exposure than the GRP companies where biomonitoring was not conducted.

## DISCUSSION

This study is the first attempt to estimate and analyse trends in the styrene exposure of workers in the European GRP industry in the period 1966–2002. These estimates are based on >300 measurements series representing >30000 samples. The styrene exposure data were retrieved from many sources including databases, reports and peer-reviewed articles published in scientific literature. A major question is of course whether our estimates reflect the actual exposure to styrene in the European GRP industry during the period 1966–2002. Various sources of bias are discussed below.

#### *Unequal distribution of exposure data*

The distribution of use of polyester resins over the European countries in 2002 is not reflected in an equal distribution of styrene exposure measurements within the regions. For example, the database contains

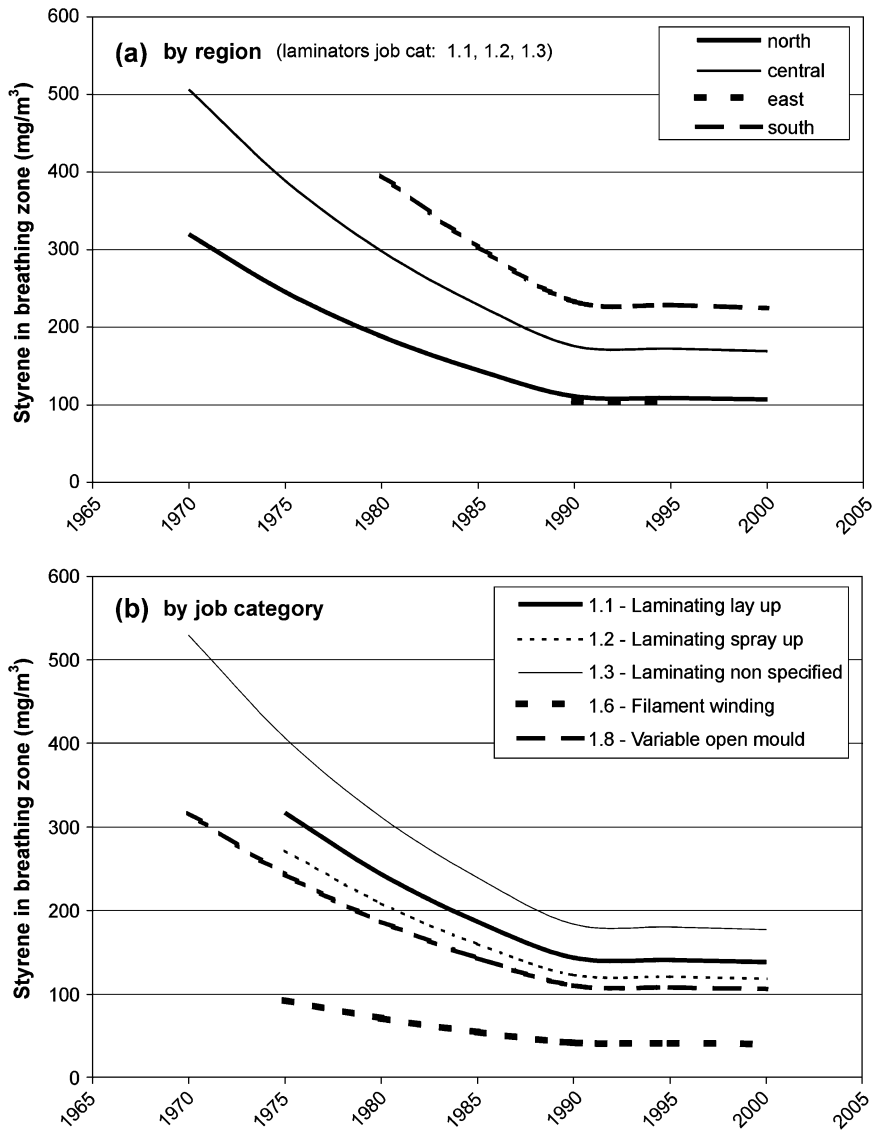


Fig. 3. Estimated temporal trend of breathing zone styrene concentrations of European open-mould workers for the period 1970–2000: by region (a) and by job category (b).

relatively many styrene exposure measurements series from the Nordic countries and relative limited exposure measurement series from Spain and Portugal.

#### Random or worst-case sampling

A substantial part of the retrieved styrene exposure data is from national databases of four European countries. There are strong indications that these databases may contain exposure data from companies and/or workers in the GRP industry with relatively high exposures.

The databases are:

1. Germany—MEGA database: Contains measurements, which were taken as a part of a supervisory

Table 4. Estimated levels of styrene in breathing zone of European open-mould process workers in 2003 (in  $\text{mg m}^{-3}$ ; 95% confidence interval)

Region	Laminators job category—1.1, 1.2, 1.3	Variable open mould—job category: 1.8	Filament winding job category—1.6
North	106 (79–143)	79 (59–105)	30 (18–51)
Central	168 (125–225)	124 (94–164)	47 (28–79)
South	222 (168–295)	165 (121–225)	63 (37–108)
All regions	141 (107–187)	105 (79–140)	40 (24–67)

Given that logarithms of the AMs have been modelled, the estimated mean represents the geometric means of the AMs. The 95 % confidence interval of estimates are presented within parentheses. Data from Eastern Europe were too limited (only 12 observations in a relative short period of 9 years (1989–1997)).



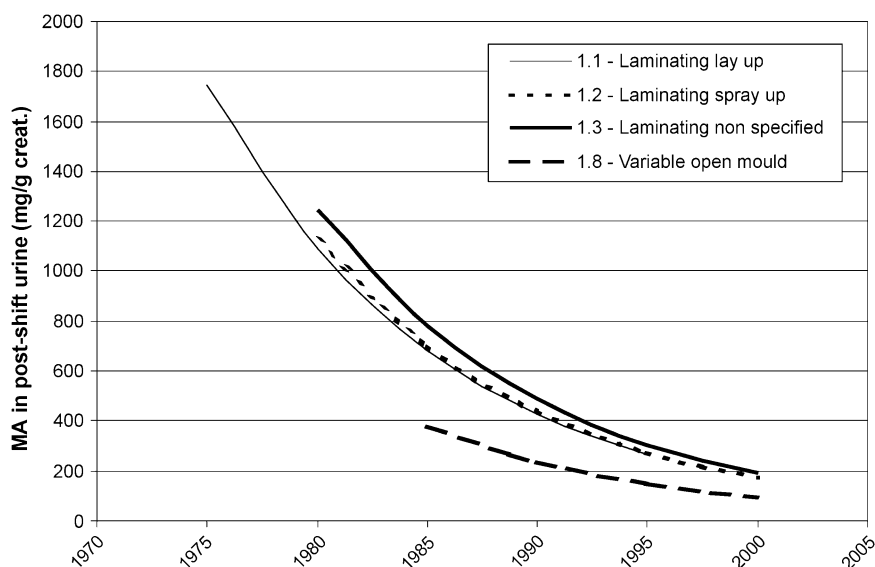


Fig. 4. Estimated temporal trend of mandelic acid in post-shift urine of European open-mould workers by job category for the period 1975–2000.

duty for various reasons and/or on the grounds of a suspected occupational disease. In cases of suspected occupational disease, these measurements are often taken in ‘worst-case’ conditions.

2. United Kingdom—National Exposure Data Base (NEDB)-HSE: Exposures reported to the NEDB generally represent the worst-case scenarios, as the monitoring work is usually undertaken as a reaction to perceived problems with a company or industry. In the draft European Risk Assessment report on Styrene (November 2007), it is stated that: ‘this can lead to a bias in the level of exposure recorded on the NEDB database’ (HSE, 2007).
3. Norway—EXPO database: Analysis of both the distribution and skewness of the styrene exposure measurements (4141 measurements) collected by the Norwegian Industry in the exposure database EXPO at the National Institute of Occupational Health in Oslo show a quasi-lognormal distribution with a negative (!) skewness (Osvall and Woldbaek, 1999). This indicates that the worst-case measurements are over-represented in the database.
4. Denmark—Danish database: Kolstad et al. (2005) report that the Danish database until 1980 was filled with results from ‘worst-case’ measurements and after 1980 with more representative measurements from a surveillance program. The authors conclude that the styrene exposure levels are possibly biased towards higher values.

In the presented analysis, ‘sampling strategy’ appeared to have no statistically significant influence on mean styrene concentrations in open-mould processing. A possible explanation is that most reports and publications contained little if any information

on the applied measurement strategy. The studies did neither report how companies were chosen nor did they contain information on how workers in these companies were selected for monitoring. The sampling strategy of all 213 mean styrene concentrations in workers applying open-mould techniques was classified as follows: ‘not reported’ (132 mean values; 62%), ‘random’ (61 mean values; 29%), ‘worst case’ (20 mean values; 9%). In addition, criteria for random and worst-case sampling were and are not well defined among researchers.

#### *Intra-individual variation in styrene exposure*

Symanski et al. (2001) showed that the intra-individual variation in styrene exposure among GRP workers can be very high in comparison to the inter-individual variation, based on results of repeated styrene measurements among workers from five different open-mould production facilities. As a consequence, a mean value that is calculated from a limited number of measurements might strongly over- or underestimate the actual average styrene exposure level in that specific job category in that period.

To reduce the potential bias of mean values from small series, the analyses used in this report included weighted analyses with the number of observations underlying each mean value as weighting unit.

#### *Classification of GRP workers in job categories*

There are no standard classes for jobs of workers in the GRP industry. In addition, many of the retrieved reports contain little information on tasks and duration of tasks performed by the sampled workers that allow a standardized classification of the retrieved exposure data.

For this report, the jobs have been classified according to the job category or task mentioned by the researchers in the report or publication. When no information on job category or tasks was available, the categories were classified simply as 'variable open mould' or 'variable closed mould'. If no information on production technique could be retrieved from the report or publication, the job category 'not reported' was assigned.

Assigning job categories based on specific tasks is difficult because GRP workers often perform variable tasks during a work shift. Workers who are described in reports or publications, as for example 'hand laminators', probably have performed other tasks during the measurements, such as gel coating, spraying and/or mould repair and finishing activities. Therefore, it cannot be excluded that some of the retrieved styrene exposure data are labelled with an inappropriate job category.

Another important question is what type of control measures mostly contributed to the decline of styrene exposure in the European GRP industry. And what caused the substantial differences in styrene exposure between the regions? Unfortunately, relevant information on, for example, the type of resin, ventilation circumstances during the measurements and actual use of personal protective equipment during monitoring was mostly not available in the publications and reports that we screened. In general, one can state that, for example, the introduction of LSE resins took place in the '70-'80s. Also the awareness of health hazards related to styrene exposure has increased in that period. Moreover, it is clear that the ambient temperature in workplaces in Southern Europe is higher than in Nordic countries. However, detailed information on the time scale of the introduction of control measures in the various European regions is lacking. Therefore, any conclusion on the contribution of control measures to the decline in styrene exposure or on the regional differences in styrene exposure remains speculative. Noteworthy is the lack of a downward trend after 1990 in the open-mould sector (only 0.4%). No apparent reason for this phenomenon could be found in the available information of the selected surveys.

## CONCLUSIONS

Trend analyses of the available styrene exposure data showed that the average styrene concentration in the breathing zone of open-mould workers in the European GRP industry has decreased on average by 5.3% per year during the period 1966–1990 and only 0.4% in the period after 1990. The highest exposures were measured in Southern Europe and the lowest exposures in Northern Europe with Central Europe in between.

Mandelic acid in post-shift urine showed a somewhat steeper decline (8.9%), most likely because

urine samples were collected in companies that also showed a steeper decline in styrene exposure concentrations in air.

Exposure data of styrene in the GRP industry retrieved for this review could have been biased towards higher values because of non-random sampling in earlier years. However, available information on measurement strategy and purpose of the measurements did not indicate this in the statistical analyses. Nevertheless, lack of detail on applied measurement strategies and purpose of surveys precluded a definite conclusion on this issue.

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## APPENDIX 1

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