

Exposure to Organic Solvents among Handicraft Car Painters: A Pilot Study in Italy

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Abstract: Car repair painters usually experience long-term exposure to many different solvents. In Italy, the greater part of car painting shops are “handicraft”, i.e. 2–5 workers and small premises. Usually workers do not have specific duties, but everyone takes part in all different operations. Moreover, working time is not standardized but varies according to the workload, working methods tend to be traditional, and compliance to individual protection devices is poor. We have hence assessed, on a sample of 8 italian handicraft car painting shops, the exposure levels to solvents, implementing three classic exposure monitoring methods: environmental sampling with charcoal tubes, personal sampling with diffusive charcoal samplers, and urinary determination of unmetabolised solvents. A simple regression analysis was performed to evaluate relationships between the three series of data. The solvents analysed were toluene, ethylbenzene, 1, 2-dichloropropane, n-butylacetate, n-amylacetate, xylene isomers, ethylacetate, and benzene. Benzene was included due to several factors, among others its presence (1% v/v) in the italian unleaded gasoline. Benzene was found in all shops, at levels around or higher than the 8-h time-weighted average limit (8-h TLV–TWA). Other solvents were found in the different shops at various levels, 10^{-2} – 10^{-1} times the 8-h TLV–TWA. Air concentrations of toluene, n-butylacetate, xylenes, and benzene were positively correlated with urinary levels of the parent compounds, while a negative correlation was found for ethylbenzene. The health implications of workers’ exposure level was briefly discussed.

Key words: Car painters, Exposure, Solvents, Benzene, Air monitoring, Urine monitoring

Introduction

Adverse health effects of organic solvents have been well known for over a century¹⁾ and have been recently described in many studies^{2–4)}. At low or moderate concentrations in air, organic solvents may cause transient symptoms such as euphoria, headache, and vertigo^{5, 6)}, while higher levels may lead to anesthesia, cardiovascular and respiratory diseases and even death³⁾. Long-term exposure may cause damage to the Central Nervous System (CNS), such as cognitive and emotional deficits⁷⁾. In particular, solvent related chronic encephalopathy (SRCE) has been described in several studies^{1, 8, 9)}. Toxic effects of organic solvents were reported to harm liver¹⁰⁾,

kidney¹¹⁾ and skin¹²⁾.

Occupational exposure to mixtures of organic solvents has been evaluated in many activities, such as painting, paint spraying, floor-laying, shoe making, laundries, graffiti removers, etc^{13–16)}. Several studies have focused on car repair painters, since this class of workers usually suffers of long-term exposure to relatively high levels of many different solvents^{2, 17, 18)}. However, in the last 10–15 years, exposure to organic solvents has been reduced for car repair painters, due to amendments to the regulations concerning collective and personal protective equipment in car painting shops, and substitution or reduction of solvents use^{9, 19)}.

This has been confirmed by recent studies carried out in Europe, which showed low exposure levels to solvents and a low frequency of acute symptoms among car

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painters during painting operations^{3, 19}). Bochelmann *et al.*²⁰ report neuro-psychological consequences, such as deficits in concentration and memory, and increase of reaction time, among car painters with long-term exposure to solvents, when compared to unexposed subjects.

In Italy, there are about 20,000 car painting shops, employing more than 80,000 workers. A few of them are classified as "industry", with 10 or more workers, while the great majority are "handicraft" with 2–5 workers and small premises, employing about 50,000 people²¹). In handicraft car painting shops, workers do not have specific duties but everyone takes part in all the different operations: surface preparation, cleansing, masking, varnish preparation, spraying, etc.

Moreover, on the basis of preliminar personal unpublished observations during several working days in some handicraft car painting shops, we highlighted some critical points:

- working time is not standardized but varies according to workload;
- working methods tend to be traditional;
- compliance to personal protective devices (PPD) is poor;
- gasoline is often used as a diluter or degreaser.

In summary, a large number of italian car painting shops employ few workers without specific duties and precise working times. Painting booths with ventilation systems are always present in the workshops, but PPD are often not used due to the lack of specific and effective training, explaining their relevance to prevent harms to workers' health: they are just considered cumbersome and slow down the work.

According to the above mentioned preliminary observations it is not possible to evaluate the exposure of a typical handicraft car painter on the basis of existing data, generally obtained in studies carried out on "industrial" car painting shops, where duties, timetables and safety rules are fixed and generally respected.

The aim of the present investigation was to assess, on a sample of 8 italian handicraft car painting shops, the exposure levels to solvents, implementing three classic exposure monitoring methods. The methods were: environmental sampling with charcoal tubes, personal sampling with diffusive charcoal samplers, and determination of unmetabolised solvents in urine. For several volatile organic compounds (VOCs) a direct relationship between urinary levels and average levels in the exposed air has been reported^{22–29}).

Materials and Methods

Eight "handicraft" car painting shops were selected in Rieti, the main town of a district in central Italy. In all

shops, operations were carried out in a large hall (200–300 m²), equipped with spray booth with ventilation system for outdoor air introduction and air ejection to outside the shop. A small office and a toilet were also present in all premises. All shops lacked a general conditioning system.

The study was performed in five steps: preliminary evaluation, air sampling, urine sampling, sample treatment and analysis, and data analysis. In each shop, one personal, one stationary, and one urine sample were collected. The study was conducted during spring period, with a mean diurnal air temperature of 21.5°C.

Step 1

A preliminary evaluation of technical data sheets of all used chemical products was performed to identify the VOCs potentially present in the workplace air. In all eight car painting shops, the identified substances were toluene (TOL), ethylbenzene (EB), 1, 2 dichloropropane (DCP), n-butylacetate (BAC), n-amylacetate (AAC), xylene isomers (XYL), ethylacetate (EAC). Although not present in the data sheets but submitted to a very restricted use, benzene (BZ) was included, due to following reasons:

- possibility of fuel vapour from cars under repair³⁰ since italian gasoline contains about 1% v/v of benzene³¹;
- observation that car repair workers often use gasoline for degreasing and diluting, an attitude developed for various reasons such as availability, economy, and ignorance of possible adverse health consequences. In general, one or more plastic bottles (about 100–150 ml) containing gasoline and a paint-brush are present in the working hall;
- possibility of the presence of small amount of BZ as an impurity in varnishes and in technical solvents used.

Step 2

Air monitoring was conducted by personal and stationary sampling. For each shop, one no-smoking painter was selected, in order to avoid benzene contribution due to cigarette smoke in urine samples. Because it was impossible to define work duration and a specific duty, both sampling methods were used at the same time, restricting the sampling period only to working steps including varnish use (preparation, spraying, tool cleaning, etc).

Personal sampling was performed by passive diffusive samplers (SKC USA, activated charcoal 350 mg), placed in the breathing zone of the worker. Environmental sampling was carried out by active sampling tubes (Supelco USA, activated charcoal 150 mg), placed 1.5 meter high and approximately in the middle of the hall, with the aspi-

ration flow fixed at 0.2 liters/minute, according to a NIOSH analytical method 1501 for aromatic hydrocarbons³²⁾ and 1450 for esters³³⁾.

Painting activities were always performed in spray booths with a ventilation system. The worker (always without a PPD) was not continuously present within the cabin, because he was also engaged in the preparation of varnishes or other activities.

Step 3

Urine samples were collected at the end of the working period, on the same day of personal and stationary sampling. The same worker selected for personal air monitoring was invited to urinate before starting air sampling, and do not urinate during it. Samples were collected in borosilicate glass bottles (250 ml), and immediately transferred into 40 mL glass vials, filling completely them up to avoid loss of analyses.

Step 4: Analysis.

Personal and stationary samples: At the end of the sampling period, samplers and tubes were stored at + 4°C and analysed within 24 h. Each active and passive sampler was desorbed with 1 mL of carbon disulphide, then analysed by gas chromatography—mass spectrometry (GC-MS): a preliminary analysis in SCAN mode, to identify potential non-targeted solvents, and a quantitative analysis in Selected Ions Monitoring (SIM) mode. Quantitative analysis was carried out by the external standard technique, preparing for each solvent, working standard solutions (w.s.s.) in carbon disulphide at different concentrations. Linearity was evaluated in the range 0.2–100 µg/ml for each solvent, always obtaining correlation coefficients greater than 0.998. The precision of the method was assessed by performing seven quality control checks for the w.s.s. at 2.0 µg/mL. The instrument lower detection limit (I-LDL), was defined as the amount giving a peak height equivalent to three times the noise level, and was calculated for each solvent on w.s.s.

Urine samples: The vials containing urine samples were stored at + 4°C, then analyzed within 24 h by using a Solid-Phase MicroExtraction (SPME) technique and gas chromatography—mass spectrometry (GC-MS) in Selected Ions Monitoring (SIM) mode, following the analytical procedure described in Guidotti and Vitali²⁴⁾.

Quantitative analysis was carried out by the external standard technique, preparing stock standard solutions in methanol for each solvent; w.s.s were prepared by spiking solvent-free urine by adding appropriate aliquots of the methanolic stock solution. Spiked urine samples, obtained from non smoking donors, were previously analysed by the same method, and target solvents always produced levels less than the limit of detection.

Linearity was evaluated in the range 0.2–100 µg/L for each solvent, always obtaining correlation coefficients greater of 0.987. The precision of the method was assessed by performing seven quality control checks for the w.s.s. at 2.0 µg/L.

The lower detection limit of the analytical procedure for urine samples (A-LDL), was defined as the amount giving a peak height equivalent to three times the noise level, was calculated on spiked urine w.s.s. for each solvent.

Step 5

Analytical data obtained from the previous steps were used for a simple regression analysis; a series of linear correlation coefficients (r) were calculated, without considering the correction for sampling duration:

- air concentrations of solvents measured by personal sampling vs air concentrations measured by stationary sampling;
- air concentrations of solvents measured by personal sampling vs urinary levels;
- air concentrations of solvents measured by stationary sampling vs urinary levels.

The PC software SPSS (SPSS, Chicago, USA) was used for the correlation analyses.

Results

Table 1 shows the analytical data obtained in the environmental monitoring (personal and stationary sampling), as well as the sampling duration, the I-LDL, and the TLV–TWA³⁴⁾. Data obtained from urinary monitoring are reported in Table 2 with their respective A-LDL. Table 3 reports a series of correlation coefficients, calculated as described in step 5.

Discussion

Air monitoring

As shown in Table 1, personal sampling produces higher values than stationary sampling, confirming that the exposure of studied car painters usually is an aggregation load of solvents, representing the sum of a background level present in the workshop plus several exposure to single substances used for specific operations.

With the exception of BZ, only solvents listed in the technical sheets of used varnishes were found in the workshops. The air levels of all solvents were in the range 0.1–22.0 mg/m³, except for shops 2 and 3, where higher levels were found, ranging from 1.8. to 93.8 mg/m³.

Car painters working in shops 2 and 3, in fact, were noted to completely ignore any precaution in varnishes manipulation, in order to accomplish their duties in a

Table 1. Data obtained from air monitoring in eight car painting workshops

Shop	Sampling duration (min)	Type of monitoring	benzene (mg/m ³)	toluene (mg/m ³)	n-butylacetate (mg/m ³)	xylenes (mg/m ³)	ethylbenzene (mg/m ³)	n-amylacetate (mg/m ³)	ethylacetate (mg/m ³)	1,2-dichloropropane (mg/m ³)
1	255	Personal	0.5	5.3	16.0	2.3	6.6	0.3	a	a
		Stationary	0.1	2.6	1.5	0.2	0.5	0.1	a	a
2	301	Personal	53.1	93.8	100.2	72.5	23.8	a	11.6	a
		Stationary	29.4	21.2	26.5	18.4	4.9	a	5.1	a
3	323	Personal	18.8	23.5	a	75.0	9.4	a	a	5.3
		Stationary	47.0	14.8	a	10.1	1.8	a	a	5.3
4	236	Personal	1.5	2.1	3.1	1.5	0.6	a	a	a
		Stationary	1.1	1.7	2.8	1.0	0.3	a	a	a
5	280	Personal	1.3	1.9	2.4	1.2	0.4	a	a	a
		Stationary	0.8	1.4	2.2	0.6	0.2	a	a	a
6	277	Personal	0.9	9.8	22.0	4.1	12.5	a	a	a
		Stationary	0.1	0.3	1.7	0.3	1.0	a	a	a
7	320	Personal	0.4	2.8	7.2	1.8	2.9	a	a	a
		Stationary	0.1	0.2	1.3	0.2	0.4	a	a	a
8	295	Personal	2.1	3.5	4.8	2.2	1.1	a	a	a
		Stationary	1.3	2.1	3.6	1.7	0.4	a	a	a
TLV-TWA ^b			1.6	188	713	434	434	266	1,440	347
I-LDL (µg/ml)			0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Target organs/effects ^b			carcinogen	CNS ^c	irritating S ^d , M ^e	irritating S ^d , M ^e	CNS ^c irritating S ^d , M ^e	irritating S ^d , M ^e	irritating S ^d , M ^e	CNS ^c , irritating S ^d , M ^e , liver, kidneys

^a<I-LDL (instrument lower detection limit), ^bACGIH, 2003, ^cCentral Nervous System, ^dSkin, ^eMucous.

Table 2. Data obtained from urine monitoring on eight car painters

Car painters	Benzene (µg/L)	Toluene (µg/L)	Butylacetate (µg/L)	Xylenes (µg/L)	Ethylbenzene (µg/L)	n-Amylacetate (µg/L)	Ethylacetate (µg/L)	1,2-dichloropropane (µg/L)
1	0.4	9.8	5.5	2.2	0.3	< A-LDL ^a	< A-LDL ^a	< A-LDL ^a
2	1.0	31.0	4.2	1.8	0.4	< A-LDL ^a	< A-LDL ^a	< A-LDL ^a
3	1.4	11.6	< A-LDL ^a	6.9	0.2	< A-LDL ^a	< A-LDL ^a	< A-LDL ^a
4	0.2	0.9	1.1	3.4	1.2	< A-LDL ^a	< A-LDL ^a	< A-LDL ^a
5	0.2	0.6	1.0	2.0	0.9	< A-LDL ^a	< A-LDL ^a	< A-LDL ^a
6	0.5	10.5	7.1	2.8	0.5	< A-LDL ^a	< A-LDL ^a	< A-LDL ^a
7	0.3	4.5	3.2	1.6	0.1	< A-LDL ^a	< A-LDL ^a	< A-LDL ^a
8	0.4	1.6	2.0	5.4	2.1	< A-LDL ^a	< A-LDL ^a	< A-LDL ^a
A-LDL ^a	0.2	0.1	0.1	0.1	0.1	0.1	0.2	0.2

^alower detection limit of the analytical procedure.

faster way. In the other shops more attention was paid to security rules even if also in these workshops the most of the safety recommendations were not followed.

BZ was found in all shops. Shops 2 and 3 (range 18.8–53.1 mg/m³) were considerably more contaminated than the others, where levels of 0.4–2.1 mg/m³ in personal sampling and 0.1–1.3 mg/m³ in stationary sampling were found. BZ values found in shops 2 and 3 were very high, even if compared with exposure levels for profes-

sional categories commonly exposed to BZ such as employees of petroleum industries³⁵, automobile mechanics³⁶, petrol pump attendants^{37–39}.

The levels found in the other shops were however 10–100 times higher than levels of BZ usually found in environmental air monitoring in Italian cities (1–50 µg/m³)^{40–43}.

BZ levels found in all shops depend only minimally on external air contamination (due to vehicular traffic), and

Table 3. Linear correlation coefficients calculated from data obtained in air and urinary monitoring

Compound	Air concentrations by stationary sampling vs air concentrations by personal sampling	Air concentrations by stationary sampling vs urinary levels	Air concentrations by personal sampling vs urinary levels
Benzene	0.71	0.97	0.67
Toluene	0.90	0.85	0.95
n-butylacetate	0.97	0.19	0.41
Xylenes	0.93	0.14	0.37
Ethylbenzene	0.93	-0.32	-0.46
n-amylacetate	1.00 (found in three shops only)	n. c. ^a	n. c. ^a
Ethylacetate	n. c. ^a	n. c. ^a	n. c. ^a
1,2-dichloropropane	n. c. ^a	n. c. ^a	n. c. ^a

^a n. c. = not calculable.

are mainly due to fuel vapour and to gasoline used for degreasing and diluting. The presence of BZ in solvents and varnishes should be verified through specific analyses.

Urine monitoring

The data obtained from urine samples agree with those obtained from air monitoring. In particular, the two workers in shops 2 and 3 show much higher urinary levels of solvents, as a result of the higher air concentrations in their workplaces.

BZ urinary levels need to be studied more in detail because non smokers are usually exposed to BZ through air, food, water and passive cigarette smoke. Consequently, the levels found in car painters' urine is the sum of occupational and daily exposure. Guidotti *et al.*⁴⁴ found an average level of urinary BZ of 0.205 µg/L (range 0.078–0.430 µg/L) in non-smoking, non-occupationally exposed subjects, and an average level of 1.593 µg/L (range 0.575–3.172 µg/L) in non-smoking petrol pump attendants; both groups were resident of Rieti, the city where the present study was conducted. Based on these data, we can consider urinary levels found in workers of shops 2 and 3 as indicators of a relevant exposure.

No parent DCP, AAC, and EAC were found in urine samples, presumably due to the fact that these solvents were present at low levels in the air, as shown in Table 1.

Assessment of health effects

The car painters involved in the study are exposed to a mixture of solvents. Air concentrations (BZ excluded) were less than their 8-h TLV-TWA in all eight shops (Table 1) and the levels were similar to those found in other studies^{3, 45}.

However, for mixtures of two or more substances that act on the same organs, it is necessary to evaluate their combined effects: several agencies (ACGIH, OSHA,

NIOSH, EPA) propose different methods to evaluate cumulative levels of exposure. ACGIH recommends an approach in which the ratio of concentration to the 8-h TLV-TWA for each component of the mixture is summed. If the sum exceeds 1, the mixture is considered to have exceeded the TLV.

We have applied this additive approach to the data obtained by personal sampling in shops 2 and 3 (higher solvent concentrations—worst cases), considering the target organs as reported in Table 1³²). All values obtained by the proposed calculation were < 1.

For BZ, however, the levels found show a worrying situation in most of the studied workshops:

- only 2 workers out of 8 are exposed to BZ levels less than 50% of the TLV-TWA (1.6 mg/m³);
- 3 workers out of 8 are exposed to BZ levels higher than TLV-TWA;
- car painters in shops 2 and 3 are exposed to concentrations 10–30 times higher than TLV-TWA.

Epidemiological data show an increased risk for non-lymphocytic leukaemia, other haematopoietic malignancies and all neoplasms^{46–49}) for long-term exposures at BZ levels comparable to those found in shops 2 and 3.

Simple regression analysis

BZ levels found in post-shift urine were positively correlated with workplace air concentrations measured by stationary ($r = 0.97$) and personal ($r = 0.67$) sampling. A closer correlation was found for stationary sampling.

Regression analysis for TOL, BAC, and XYL showed a positive correlation for urine levels both with stationary and personal air monitoring; this correlation was stronger for TOL ($r = 0.85$ and $r = 0.95$ respectively), confirming a previous study by Takeuchi *et al.*²⁸), and higher for personal than stationary sampling for all three solvents. Car painters exposure to these VOCs was mainly due to use of varnish.

EB shows a negative correlation with both air sampling

methods. On the contrary, several studies^{25, 28)} have found a positive correlation between EB exposure concentrations and urinary levels in humans, although this correlation was weaker than other VOCs.

Environmental and urinary data on DCP, AAC, and EAC were insufficient to apply a regression analysis.

Conclusions

In conclusion, Italian handicraft car painting shops workers are exposed to a mixture of solvents commonly used in varnishing, and to benzene, due to fuel vapour from cars in repair and to the use of gasoline for degreasing and dilution. The exposure to solvents derives mainly from the application of paints, as it is shown by the comparison between stationary sampling and personal sampling, even if the differences in concentration are not large. In any case, the values found were lower than the corresponding 8-h TLV-TWA, both for single solvents and for their mixture.

On the contrary, exposure to benzene is high in all workshops, ranging from 25% to 300% of the 8-h TLV-TWA.

From a general point of view, the need to implement preventive activities for handicraft car painters in Italy is evident:

- monitoring activities should be extended to a larger number of handicraft car painting shops, in order to obtain data from a more appropriate sample. Monitoring should include both air and urinary analyses;
- there is a need for a better control on exposure to solvents, to be realized by means of improvement and development of ventilation systems and enforcing personal respiratory protection for high intensity exposure tasks;
- workers in these environments should be made aware of the potential risks by means of information and training;
- workers should be forced to use, at least during critical working phases, respiratory protection devices.

As for benzene, considering that this compound is not declared in the technical sheets of the used paints and it is subject to very restricted use due to its toxicity, it is our opinion that:

- it is necessary to reduce the time of parking of cars inside the workshop;
- there is a need for a specific training on health risks related to benzene exposure and on improper use of gasoline;
- it is necessary to periodically monitor solvents exposure in car painting shops, especially benzene levels. It could be useful to measure levels of aliphatic

hydrocarbons, such as nonane and octane, in order to control the presence of gasoline vapour sources;

- it could be useful to analyse technical solvents and varnishes commonly used in car painting, to reveal the possible presence of BZ.

These activities should be coordinated and managed at a national level by car painters trade unions or associations.

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