Organic Solvent Exposure in Small Scale Industries in North-East Japan

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(Received March, 11, 1985 and in revised form June 8, 1985)

Abstract: Exposure to organic solvent vapors was investigated in 40 unit workplaces (with 189 workers) in 16 small-scale industries in north-east Japan (Tohoku District) in which synthetic *urushi* lacquer was applied to produce non-metal tableware (e.g. soup bowls, rice bowls), flower vases, home altars, etc. Two furniture factories were also studied. The equipment used was carbon felt dosimeters and a portable PID-GC (Model 10A10, Photovac, Ontario, Canada; with minor alterations). Under the conditions employed, the GC could analyze benzene-toluene-xylenes within 150 seconds. Independent of the sizes of the factories studied, efficient local exhaust systems were very popular in spray painting rooms. Toluene was the major pollutant in the workplace air, with small quantities of xylenes, ethylbenzene, and much less frequently, n-hexane. Exposure did not exceed the current occupational exposure limit in all the cases except for the two workers, who were excessively exposed due to the generation of dense vapors in automated spraying process. In contrast, the maintenance of drying process was rather poor. In the cases of the production of high quality goods, no artificial ventilation system was allowed in afraid of pollution of finished surface with fine dust, and the vapor levels in the workroom tended to be higher towards the end of the shift.

Key words: Carbon felt dosimeter—n-hexane—organic solvent—portable PID-GC—small scale industry—toluene—*urushi*-ware—xylenes

This work was supported by a Grant-in-Aid for Co-operative Research (No. 58370017) for 1983-1984 from the Ministry of Education, Science and Culture of the Government of Japan.

A part of the work was presented at the 58th Annual Meeting of Japan Association of Industrial Health held in Kitakyushu on 27-30, March, 1985.

INTRODUCTION

Organic solvents are quite common materials in various types of industries, and they have been introduced even in very traditional factories like *urushi*-ware (Japan-lacquerware) workshops, as the solvents in synthetic *urushi* paints. While solvents use in modern industries have been extensively studied and exposures of workers were also well documented,¹⁻⁵⁾ less attention has been given to analyze solvent exposure condition in family labor-supported cottage industries. Because north-eastern part of Japan, especially Tohoku District, is known to be rich in such industries, survey was initiated as a regional co-operative research to elucidate the intensity of exposure of workers in small-scale factories manufacturing lacquerware such as soup bowls, rice bowls, flower vases, tables or even Buddhistic home altars with synthetic *urushi* paints. Furniture industries were also investigated where solvent-containing synthetic glue was used.

MATERIALS AND METHODS

In total, 16 factories of various sizes in 5 prefectures in Tohoku District were studied in 1983–1985. The major products were non-metal tableware (i.e., rice bowls, soup bowls, etc.), Buddhistic home altars and other miscellaneous *urushi* ware (e.g., flower vases, service trays, letter boxes). Two furniture factories were also included. In these factories, 40 unit workplaces were investigated for the environmental concentration (Measurement A, taking grid-sampling strategy⁶⁾) and also for the possible maximum vapor concentration in the breathzone air of workers therein (Measurement B⁶⁾). Individual exposure concentrations were measured with 189 workers serving there.

Average exposure concentrations and environmental concentrations (Measurement A with grid sampling strategy) for an entire workday, e.g., 9:00 a.m. to 5:00 p.m., were measured taking advantage of carbon felt dosimeters.⁷⁻⁹⁾ After the exposure was terminated, the felt was stored in a refrigerator¹⁰⁾ for a week at maximum, and extracted with carbon disulfide (with 240 μ l sec.-butylbenzene/L as an internal standard). An aliquot of the extract was then subjected to FID-GC analysis. Measurement B was carried out by the injection of 0.25–1.00 ml of the grab-sampled air in the breathzone of workers into a portable photo-ionization detector-gaschromatograph (Model 10A10, Photovac, Ontario, Canada). As a preliminary study revealed that the major components of the solvents in use were toluene, xylenes, ethylbenzene, and less frequently, n-hexane, a polytetrafluoroethylene (Teflon) column (30 cm in length and 2.5 mm in inner diameter) packed with Carbowax BHT-100 (40–60 mesh) was employed. The supply of the carrier gas, clean air, was at ca. 2.8 kg/cm² (40 psi). This equipment is, by its structure, not explosion-inducing because it does not use any flame. For the survey purpose, the equipment was further altered as follows: (1) An extension

switch was installed to reduce the detector sensitivity by 1/100 so that aromatics at ppm levels could be measured when 1 ml of the sample air was injected. (2) In a preliminary study, it was found that the baseline of the chromatogram drifts as a function of the room temperature, as the GC originally does not have any heating system for the column. Accordingly, a heating system was placed by fixing a heater plate (Thermicon-heater Model K; Sakaguchi Electroheater Co., Tokyo) on the outside of the metal box which accommodated the column. The heater plate was connected with a thermo-sensor (Model RO 60-13; Chino Co., Tokyo) via a voltage regulator (Model: Electro-slider, Iuchi-Seieido Co., Osaka) and a thermo-controller (Model R 7650B, Yamatake-Honneywell Co., Tokyo) as shown in Fig. 1. Although the system can be adjusted to a range of temperature from the room temperature to 50°C, it was operated in the present study to keep the air inside the column box at 30°C. (3) The storage batteries set in the bottom of the GC case was removed to reduce the weight to a final of 10 kg. One analysis by the GC could be achieved in 3 minutes. For the identification of solvents especially xylene isomers, capillary GC was employed under the conditions previously described.^{11,12)}

For the preparation of standard solvent vapor at given concentrations, the servo-mechanized vapor generation system¹³⁾ was used.

In order to examine the reproducibility of the analyses with the portable GC, the determinations of toluene in air were repeated 5 series a day for 5 days, each series being composed of 3 determinations each at 50 and 100 ppm; the detector

Fig. 1. Installation of a heating system in the PID-GC.

- A: a heater plate.
- B: a voltage regulator.
- C: a thermo-controller.
- D: a thermo-sensor.

was switched on anew at the beginning of each series. There was no change in the toluene chromatograms during the 5 day examination period.

As more than one solvent was usually detected, the sum in the additiveness formula^{14,15)} (i.e., $\sum_{i=1}^n \frac{Ci}{OELi}$ where Ci and $OELi$ are the observed concentrations of the solvent i and the corresponding occupational exposure limit, respectively) was employed to make overall evaluation of the multi-component mixture of the solvent vapors. The occupational exposure limit cited was 100 ppm each for toluene, xylenes, ethylbenzene and n-hexane.¹⁶⁾ When 3 or more determinations were made either for exposure concentrations or environmental concentrations, the results were expressed in terms of the geometric mean followed by the geometric standard deviation in the parenthesis and the number of determination.

RESULTS AND DISCUSSION

The typical process to paint *urushi* ware with synthetic *urushi* lacquer was composed of 5 to 6 steps. The material (originally wooden, but more popularly plastic one already in the shape of, e.g., bowl or vase etc.) was first undercoated. After the undercoat dried, the coating followed by drying was repeated as necessary until the final coating. When requested, the coat may be polished with sand-paper or whetstones to smoothen the surface or to obtain specified pattern after the final coating. Sometimes, additional painting with brushes, a mimeographic technique or else would be made over the final coat to finish the process. In the case of home altar production, plain wood pieces were put together with glue to make shapes prior to undercoating.

While toluene was the major contaminant of the workroom air, xylenes (three isomers of xylene and ethylbenzene) were present in most cases (i.e., all the workplaces except for Cases 29–32 and 35) presumably as impurities (5–20% of toluene, as total xylenes), and n-hexane was also detected in Cases 5, 6 and 8–10 (concentration being 10–30% of that of toluene).

The results of the survey are summarized in Table 1 by the products in the workplaces and by the sizes of the factories.

Worthy of noting is the fact that the geometric mean exposure concentration was less than 1 unit or, in other words, were below the current occupational exposure limit in almost all the workplaces studied, except for Case 6. Correspondingly, the levels by Measurements A and B were also low. Such low levels were primarily achieved by popular use of the local exhaust booth and a powerful fan to remove the dispersed droplets. The installation of the exhaust system was apparently not limited to the large-scale industries, but even a very small family-based factory used to have an effective system, although the system usually lacked any solvent scrubber to prevent outdoor air pollution. Thus, the exposure intensity did not vary remarkably among the factories of either different sizes nor of different products. The variation in exposure intensity among various jobs was

Table 1. Exposure and environmental concentrations in various *urushi* ware and furniture workplaces

Products	Cases	Sizes ^a	Solvent work ^b	Exposure concentrations ^{c,d}	Environmental concentrations ^{c,e}	
					Measurement A	Measurement B
Bowls ^f	1	F	Hand-spray I	0.12, 0.16	0.29(1.54) : 6	0.28
	2	F	Hand-spray II	0.12	0.27(1.62) : 5	—
	3	F	Preparatory	—	0.23(1.96) : 6	—
	4	S	Brush painting I	0.04	0.02(1.25) : 4	0.03
	5	S	Hand-spray I	—	0.33(1.15) : 6	0.10
	6	S	Automated spray I	1.36(1.41) : 4 [0.96, 2.92] ^j	0.45(1.73) : 4	—
	7	S	<i>ibid.</i>	0.35(1.40) : 5 [0.24, 0.54]	0.34(1.54) : 5	0.56
	8	S	Brush painting II	0.42	0.36(1.13) : 5	0.16
	9	S	<i>ibid.</i>	—	0.12(1.65) : 6	—
	10	S	Hand-spray II	0.32(1.38) : 4 [0.26, 0.42]	0.38(2.00) : 4	—
	11	S	Screen-painting	0.44(1.59) : 12 [0.15, 0.70]	0.50(1.22) : 3	—
	12	S	Finish	0.33(1.18) : 4 [0.28, 0.38]	0.26(1.39) : 5	—
	13	S	Preparatory/Finish	0.18	0.26(1.46) : 6	—
	14	S	<i>ibid.</i>	0.26, 0.29	0.12(1.47) : 5	—
Miscellaneous ^f	15	S	Brush painting I	0.09, 0.13	0.12(1.32) : 5	0.09
	16	M	Hand-spray I ^h	0.03(1.77) : 3 [0.02, 0.05]	0.03(2.63) : 7	0.02
	17	M	<i>ibid.</i> ⁱ	0.09(2.16) : 3 [0.06, 0.12]	0.04(1.65) : 7	0.17
	18	M	Hand-spray II ^h	0.15, 0.21	0.07(1.65) : 5	0.15
	19	M	<i>ibid.</i> ⁱ	0.13, 0.31	0.08(1.93) : 5	0.22
	20	M	<i>ibid.</i>	0.02	0.06(2.06) : 5	0.25
	21	M	Screen painting ^h	0.11, 0.34	0.12(1.25) : 5	0.30
	22	M	<i>ibid.</i> ⁱ	0.11	0.03(1.60) : 5	0.13
	23	M	<i>ibid.</i>	0.15, 0.15	0.09(1.27) : 5	0.15
	24	M	Polishing ^h	0.08(1.67) : 6 [0.06, 0.15]	0.02(3.93) : 6	—
Home altars ^f	25	M	<i>ibid.</i> ⁱ	0.09(2.16) : 6 [0.03, 0.23]	0.04(1.59) : 6	0.06
	26	F	Hand-spray	0.08, 0.12	0.04(1.24) : 5	—
	27	F	Drying	—	0.12(1.68) : 5	—
	28	F	<i>ibid.</i>	0.15	0.02(1.41) : 3	0.19
	29	F	Polishing	0.13, 0.16	0.09(1.59) : 4	—

Table 1. (Continued)

Products	Cases	Sizes ^a	Solvent work ^b	Exposure concentrations ^{c,d}	Environmental concentrations ^{e,e}	
					Measurement A	Measurement B
Furniture ^g	30	F	<i>ibid.</i>	0.14(1.08) : 3 [0.07, 0.15]	0.03(1.23) : 5	0.23
	31	M	Hand-spray	0.30(1.02) : 3 [0.26, 0.37]	0.18(1.34) : 5	—
	32	M	Inspection	0.19(1.24) : 4 [0.14, 0.28]	0.17(1.28) : 4	—
	33	L	Hand-spray	0.13(2.19) : 8 [0.05, 0.27]	0.09(1.46) : 5	0.07
	34	L	<i>ibid.</i>	0.22(1.38) : 10 [0.13, 0.40]	0.27(1.56) : 5	—
	35	L	<i>ibid.</i>	0.23(1.59) : 21 [0.09, 0.75]	0.33(1.28) : 8	—
	36	L	Gluing	0.21(1.46) : 6 [0.11, 0.48]	0.29(1.38) : 8	—
	37	L	<i>ibid.</i>	0.35(1.27) : 4 [0.22, 0.43]	0.32(1.31) : 5	—
	38	L	Hand-spray	0.10(2.19) : 28 [0.03, 0.40]	0.02(1.66) : 7	0.18
	39	L	<i>ibid.</i>	0.09(2.32) : 19 [0.02, 0.58]	0.03(2.08) : 7	0.05
	40	L	<i>ibid.</i>	0.34(1.66) : 11 [0.11, 0.63]	0.27(1.46) : 9	—

^a Sizes of factories were classified by the number of the workers as;

F: essentially family members only. S: small, i.e., less than 10 workers.

M: medium, i.e., 10 to 50 workers. L: large, i.e., over 50 workers.

^b Spray (or painting) I: Undercoating.

Spray (or painting) II: Final coating.

Screen painting: Additional painting with a mimeographic technique.

Finish: Additional painting with writing brushes, etc.

Polishing: Polishing of the coated surface with sand-paper or whetstones. No solvent was used.

Inspection: No solvent was used.

Preparatory work: No solvent was used.

^c The additiveness formula^{14,15} was employed. Unit: Occupational exposure limit.¹⁶ For details, see MATERIALS AND METHODS.

^d Numbers are either individual values or geometric mean (geometric standard deviation): number of determinations (i.e., number of workers examined or number of stationary sampling sites). The minimum and maximum values are shown in square parentheses in case three or more workers were studied.

^e Geometric mean (geometric standard deviation): number of determinations.

^f Urushi ware.

^g No urushi lacquer was applied.

^{h,i} These workplaces were examined once in summer (Cases 16, 18, 21 and 24 indicated by h) and then in the following winter (Cases 17, 19, 22 and 25 indicated by i). The difference between the pairs were statistically insignificant ($p > 0.10$).

^j Two cases exceeded the occupational exposure limit.

Fig. 2. Typical distribution of solvent vapors in family-based factories.

Note wide-spread distribution even in the preparatory room (Case A) where no solvent was applied. In the brush painting rooms (Case C), no exhaust fan was installed. In Case B, the concentrations in the preparatory/finish room were almost as high as those in the hand-spray room. Unit: Occupational exposure limit.¹⁶ The additiveness formula^{14,15} was employed. For details, see MATERIALS AND METHODS.

not marked; the exposure levels of painters were not always higher than the levels of those with indirect exposures (e.g., Case 26 in comparison with Case 29). Even hand-spray painters in the smallest factories (Cases 1 and 2) were not exposed to organic solvent vapors above the current occupational exposure limit. In fact, none of the workers except for the two in Case 6 was excessively exposed to the solvent vapor. When the same workplaces were examined once in summer and then in the following winter, the comparison of the results disclosed that the vapor levels were tended to be higher in winter when the windows were closed (Cases 17, 19, 22 and 25) than in summer when the windows were kept open (Cases 16, 18, 21 and 24).

A general tendency was observed that the exposure concentration would be higher than the level the environmental concentrations suggest when the painters make very fine work, keeping their faces close to the objects [e.g., Cases 8 (final brush painting), 12 (finish) and 23 (screen-painting)], as previously described.^{6,8)} In Case 6 where geometric mean exposure concentration exceeded the limit, the two (who were over-exposed) of the four workers took the fixed positions close to spray guns to monitor the spraying operation and remaining two engaged in carrying out paint-wet articles; during the carriage, the wet articles were kept rather close to their chests.

In the case of production of very high quality goods, pollution of the finished surface by house dust was intensively afraid of, and often no artificial ventilation system was installed in the workroom (Case 8 in Table 1; also Fig. 2-C). The exposure of the worker therein was rather high and vapor levels in the workroom tended to be higher towards the end of the shift (data not shown). In case 8, the measurement B could be made only in the morning and might not reflect the highest level in the breathzone air in the afternoon.

The current major difficulties appeared to be in the drying process. In limited cases (e.g., Case 27), a room was reserved for drying process and no human exposure would take place in association with the drying process except for during the carrying-in and carrying-out. In most cases, however, there was no specified room for drying even in large-scale factories and sprayed wet materials were left in various corners in the painting room and then transferred to other preparatory rooms while they were not dry yet. Such practise brought pollution sources into other rooms (e.g., the inspection room in Case 32 or the polishing room in Case 25 where no solvent was used) and finally pollution may spread throughout the factory (Fig. 2-B and C). It is apparently desirable to establish a specified drying room to isolate the wet materials and to prevent unnecessary human exposure. The mixing of solvent-containing materials to prepare requested spray-paint could also be a source of high exposure when done carelessly. In one observed case, the mixing practise away from the exhaust booth resulted in the generation of 63 ppm toluene-xylene vapor in the breathzone of the engaged worker.

Because of limitation in survey conditions, no health examination of the workers could be conducted in the present study. The experiences through the survey, however, suggest that the irritation of the nose and the eyes felt in limited workplaces (e.g., in Case 7) cannot be explained by the exposure to the solvents at the observed levels. The possible exposure to an irritant chemical such as toluene diisocyanate in *urushi* ware production should be considered and deserves further study.¹⁷⁾

REFERENCES

- 1) WHO (1981). Recommended health-based limits in occupational exposure to selected organic solvents. Technical Report Series 664, WHO, Geneva.
- 2) IPCS. *Environmental Health Criteria* Toluene, in press.
- 3) IPCS (1983). *Environmental Health Criteria* 26 Styrene, WHO, Geneva.
- 4) IPCS. *Environmental Health Criteria* Trichloroethylene, in press.
- 5) IPCS (1984). *Environmental Health Criteria* 31 Tetrachloroethylene, WHO, Geneva.
- 6) Ikeda, M. and Ohtsuki, T. (1985). Exposure concentration versus environmental concentration: A field study in organic solvent workplaces. *Tohoku J. Exp. Med.*, **146**, 225.
- 7) Hirayama, T. and Ikeda, M. (1979). Applicability of carbon felt to the dosimetry of solvent vapor mixture. *Am. Ind. Hyg. Ass. J.* **40**, 1091.
- 8) Koizumi, A., Hirayama, T., Shiojima, S. and Ikeda, M. (1980). Evaluation of carbon felt dosimetry in comparison with one-shot air sampling method and biological monitoring: A field survey. *Ind. Health*, **18**, 153.
- 9) Hasegawa, K., Shiojima, S., Koizumi, A. and Ikeda, M. (1983). Hippuric acid and *o*-cresol in the urine of workers exposed to toluene. *Int. Arch. Occup. Environ. Health*, **52**, 197.
- 10) Ikeda, M., Kumai, M. and Aksoy, M. (1984). Application of carbon felt dosimetry to field studies distant from analytical laboratory. *Ind. Health*, **22**, 53.
- 11) Kumai, M., Koizumi, A., Saito, K., Sakurai, M., Inoue, T., Takeuchi, Y., Hara, I., Ogata, M., Matsushita T. and Ikeda, M. (1983). A nationwide survey on organic solvent components in various solvent products: Part 2. Heterogeneous products such as paints, inks and adhesives. *Ind. Health*, **21**, 185.
- 12) Ikeda, M., Kumai, M., Watanabe, T. and Fujita, T. (1984). Aromatic and other contents in automobile gasoline in Japan. *Ind. Health*, **22**, 235.
- 13) Koizumi, A. and Ikeda, M. (1981). A servomechanism for vapor concentration control in exposure chambers. *Am. Ind. Hyg. Ass. J.*, **42**, 417.
- 14) Elkins, H. E. (1962). Maximum allowable concentration of mixtures. *Am. Ind. Hyg. Ass. J.*, **23**, 132.
- 15) ACGIH (1984). Threshold limit values for chemical substances and physical agents in the work environment and biological exposure indices with intended changes for 1984–85. p. 47. ACGIH, Cincinnati, OH.
- 16) Japan Association of Industrial Health (1984). Recommended occupational exposure limits. *Jpn. J. Ind. Health*, **26**, 339.
- 17) Hattori, M. (1984). Respiratory symptoms in lacquer ware workers. *Jpn. J. Hyg.*, **39**, 429 (in Japanese).