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Organic Solvent Use in Enterprises in Japan

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Abstract: This study was initiated to elucidate possible changes in types of organic solvents (to be called solvents in short) used in enterprises in Japan through comparison of current solvent types with historical data since 1983. To investigate current situation in solvent use in enterprises, surveys were conducted during one year of 2009 to 2010. In total, workroom air samples in 1,497 unit workplaces with solvent use were analyzed in accordance with regulatory requirements. Typical use pattern of solvents was as mixtures, accounting for >70% of cases. Adhesives spreading (followed by adhesion) was relatively common in small-scale enterprises, whereas printing and painting work was more common in middle-scale ones, and solvent use for testing and research purpose was basically in large-scaled enterprises. Through-out printing, painting, surface coating and adhesive application, toluene was most common (being detected in 49 to 82% of workplaces depending on work types), whereas isopropyl alcohol was most common (49%) in degreasing, cleaning and wiping workplaces. Other commonly used solvents were methyl alcohol, ethyl acetate and acetone (33 to 37%). Comparison with historical data in Japan and literature-retrieved data outside of Japan all agreed with the observation that toluene is the most commonly used solvent. Application of trichloroethylene and 1,1,1-trichloroethane, once common in 1980s, has ceased to exist in recent years.

Key words: Annual trend, Isopropyl alcohol, Organic solvent, Solvent mixture, Toluene

Introduction

Organic solvents (to be called solvent in short) are among the most common industrial chemicals, and have been used in various corners in industries both in developed and developing countries. Types of solvents in use may shift from one to another in reflection of numerous factors such as growth and decay of the industries[1–3] as well as occuoational health concerns[4–6] and consideration for green chemistry[7, 8]. In case of Japan for example, new cases of occupational poisoning induced by exposure to carbon disulfide[1], benzene (then a glue solvent[2]), or *n*-hexane[3] are no longer reported as rayon industries have ceased to exist and cites of production of synthetic material-based foot wares have moved to other countries. Thus, continuous monitoring for types of solvents commonly used in industries is an important issue for proper management of occupational health in solvent workplaces.

This study group has been compiling data on solvent components commonly detected in various workplaces in enterprises since 1980s[9–14]. The historical data will be compared in the present report with the results of the latest air analysis to clarify trends in the types of solvents in solvent workplaces. Results of similar analyses by this group for solvent use in research institutions have been reported separately[15].

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Table 1. Organic solvents registered in the Regulations for Prevention of Organic Solvent Poisoning

Group 1 organic solvents (7 solvents)
- 1,2-Dichloroethane (Etylene dichloride)
- 1,2-Dichloroethylene (Acetylene dichloride)
- Carbon disulfide
- Carbon tetrachloride
- Chloroform
- 1,1,2,2-Tetrachloroethane (Acetylene tetrachloride)
- Trichloroethylene

Group 2 organic solvents (40 solvents)
- Acetone
- 1-Butanol
- 2-Butanol
- *n*-Butyl acetate
- Chlorobenzene
- Cresol
- Cyclohexanol
- Cyclohexanone
- *o*-Dichlorobenzene
- Dichloromethane (Methylene dichloride)
- 1,4-Dioxane
- Ethylene glycol mono-*n*-butyl ether (Butyl cellosolve)
- Ethylene glycol monoethyl ether (Cellosolve)
- Ethylene glycol monoethyl ether acetate (Cellosolve acetate)
- Ethylene glycol monomethyl ether (Methyl cellosolve)
- Ethyl acetate
- Ethyl ether
- Hexane (n-Hexane, so-called)
- Isobutyl acetate
- Isobutyl alcohol
- Isopentyl acetate (Isoamyl acetate)
- Isopentyl alcohol (Isoamyl alcohol)
- Isopropyl acetate
- Isopropyl alcohol
- Methyl alcohol
- Methyl *n*-butyl ketone
- Methyl acetate
- Methyl ethyl ketone
- Methyl isobutyl ketone
- Methylcyclohexanol
- Methylcyclohexanone
- *N,N*-Dimethylformamide
- *n*-Pentyl acetate (*n*-Amyl acetate)
- *n*-Propyl acetate
- Styrene
- Tetrachloroethylene (Perchloroethylene)
- Tetrahydrofuran
- Toluene
- 1,1,1-Trichloroethane
- Xylenes

Group 3 organic solvents (7 solvents)
- Coal tar naphtha
- Mineral spirit (including mineral thinner, mineral turpentine oil, petroleum spirit and white spirit)
- Petroleum benzine
- Petroleum ether
- Petroleum naphtha
- Turpentine oil

Mixtures of only these solvents

Materials and Methods

Enterprises and solvent workplaces surveyed

Surveys were conducted for one year period from April, 2009 to March 2010. In total, 1,497 unit workplaces (unit workplace as defined by the regulation[16–18]) with solvent use were examined.

Analysis for solvent vapors in workroom air

The target solvents were Group 1 (7 types) and Group 2 (40 types) solvents as prescribed in the Ordinance for Prevention of Organic Solvent Poisoning (Table 1[19, 20]), similar to the previous publications[11–14]; Group 3 solvents were not measured as they are natural solvent mixtures[11, 14]. The methods of workroom air sampling (by the Tedlar bag method) and instrumental analyses (by capillary gas-chromatography) were in accordance with regulatory standards, as previously described in details[11, 14].

Statistical analyses

χ^2 test and Cochran-Armitage's trend test were applied as necessary.

Results

Number of solvent workplaces surveyed by size of enterprises

When the 1,497 unit workplaces surveyed were classified by the number of workers employed (Table 2), a majority (651 workplaces or 43.5%) of unit workplaces belonged to medium-scale enterprises with 51 to 300 employees. This was followed by large-scale enterprises (535 workplaces or 35.7%) with 500 or more employees. Workplaces in small-scale enterprises (with ≤50 employees) counted 223 places or 14.9% of the total.

Classification by type of solvent work (Table 2) showed that one third (30.2%) of solvent use was for degreasing, cleaning or wiping, of which a half was in large-scale enterprises. This was followed by solvent use for testing and research (321 workplaces) conducted primarily in large-scale enterprises (62.9%) and very few (7.2%) in small-scale ones. The third and forth common uses were for painting (12.2%) and printing (10.3%), both being common in middle-scale enterprises (50% or higher).

Common solvents by type of work

Five most common types of solvent work, i.e.,

Table 2. Number of unit workplaces by type of solvent work and by enterprize size

Solvent work	Enterprise size by number of employees[a]														
	A (1–50)			B (51–300)			C (301–500)			D (≥501)			Total		
	No.	%[b]	%[c]	No.	%[b]	%[c]	No.	%[b]	%[c]	No.	%[b]	%[c]	No.	%[b]	%[c]
1. Production[d]	3	1.3	4.9	50	7.7	82.0	2	2.3	3.3	6	1.1	9.8	61	4.1	100.0
2. Use of solvents[e]	12	5.4	17.6	48	7.4	70.6	2	2.3	2.9	6	1.1	8.8	68	4.5	100.0
3. Printing	28	12.6	18.2	100	15.4	64.9	2	2.3	1.3	24	4.5	15.6	154	10.3	100.0
4. Handwriting, drawing	2	0.9	50.0	0	0.0	0.0	0	0.0	0.0	2	0.4	50.0	4	0.3	100.0
5. Surface coating	35	15.7	30.4	56	8.6	48.7	4	4.5	3.5	20	3.7	17.4	115	7.7	100.0
6. Adhessive spreading	27	12.1	27.0	40	6.1	40.0	14	15.9	14.0	19	3.6	19.0	100	6.7	100.0
7. Adhesion	5	2.2	45.5	5	0.8	45.5	1	1.1	9.1	0	0.0	0.0	11	0.7	100.0
8. Degreasing, cleaning, wiping	41	18.4	9.1	155	23.8	34.3	30	34.1	6.6	226	42.2	50.0	452	30.2	100.0
9. Painting	46	20.6	25.3	90	13.8	49.5	16	18.2	8.8	30	5.6	16.5	182	12.2	100.0
10. Solvent-drying	1	0.4	3.4	25	3.8	86.2	3	3.4	10.3	0	0.0	0.0	29	1.9	100.0
11. Testing, research	23	10.3	7.2	82	12.6	25.5	14	15.9	4.4	202	37.8	62.9	321	21.4	100.0
Total	223	100.0	14.9	651	100.0	43.5	88	100.0	5.9	535	100.0	35.7	1,497	100.0	100.0

[a]The number in parentheses is the number of employees in an enterprize.
[b]Percentage in the vertical direction.
[c]Percentage in the horizontal direction.
[d]Production of solvent-containing preparations.
[e]Use of solvents for solvent-containing preparations such as pesticides, pigments etc.

Table 3. Type of solvents commonly used in various solvent work

Order[b]	Type of solvent work[a]				
	3. Printing	5. Surface coating	6. Adhesive spreading and 7. Adhesion	8. Degreasing, cleaning and wiping	9. Painting
1	Toluene (64%)	Toluene (82%)	Toluene (49%)	Isopropyl alcohol (49%)	Toluene (82%)
2	Methyl ethyl ketone (62%)	Methyl alcohol (76%)	Ethyl acetate (45%)	Acetone (34%)	Xylenes (77%)
3	Ethyl acetate (59%)	Isopropyl alcohol (64%)	Acetone (32%)	Toluene (31%)	*n*-Butyl acetate (65%)
4	Isopropyl alcohol (53%)	Xylenes (54%)	Methyl ethyl ketone (26%)	Methyl alcohol (22%)	Ethyl acetate (53%)
5	Methyl alcohol (45%)	Methyl ethyl ketone (28%)	Methyl alcohol (24%)	Ethyl acetate (20%)	1-Butanol (42%)

[a]The number in parentheses indicates the percentage of the prevalence.
[b]Order of detection prevalence.

3. Printing, 5. Surface coating, 6. Adhesive spreading + 7. Adhesion, 8. Degreasing, cleaning and wiping, and 9. Printing, were selected, and types of solvents detected in these workplaces were tabulated in the order of % prevalence of detection (Table 3). It was clear that toluene was most common in 4 types of workplaces (49 to 82%). One exception was workplaces for 8. Degreasing etc., where isopropyl alcohol was the most common solvent (49%). It should be noted that, in many cases, solvents were used as mixed; the percentage shows the prevalence in the detection and not the proportion in mixing. The presence of xylenes (i.e., xylene isomers) assumedly does not mean the intentional addition of xylenes but the presence as impurities in industrial toluene. Other solvents detected were alcohols (typically methanol), ketones (especially methyl ethyl ketone) and esters (e.g., ethyl acetate).

Use of solvent mixtures rather than unmixed single solvent

As stated above, solvents were generally used as mixed. Quantitative analysis showed (Fig. 1) that the use of solvent as unmixed accounted only for 27.5% of total use (n=1,497). Thus, solvents were used as mixed in a majority (72.5%) of cases. The observation during workplace rounds was also in support of this consideration, i.e., the preparations containing multiple solvents used in the workplaces were usually made up in other places prior to the use. The largest number for solvents detected in air of a single unit workplace was 14. Although the distribution was apparently skewed, a simple arithmetic mean for the number of solvents was 3.56, and the median was 5.

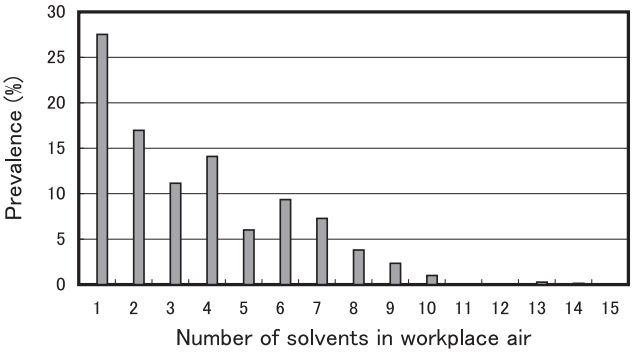

Fig. 1. Number of solvents detected in workplace air samples.
Note that the distribution does not follow a normal distribution. The median is 5 solvents, whereas a tentative arithmetic mean is 3.6 solvents/air sample.

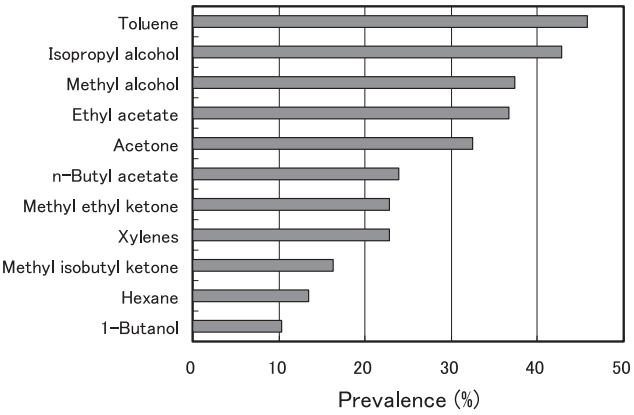

Fig. 2. Top eleven solvents of highest prevalence in use.
Those with ≥10% prevalence (11 solvents) are selected.

Commonly used solvents

Figure 2 shows the solvents detected in 10% or more of the 1,497 workroom air samples. In practice, they were 11 in total, and toluene was most common among them, being detected in 46% of the total samples. This was followed by isopropyl alcohol (42.8%). Other solvents were two alcohols [methanol (37.8%) and 1-butanol (10.3%)], two acetate esters [ethyl acetate (36.7%) and *n*-butyl acetate (23.9%)], and three ketones [acetone (32.5%), methyl ethyl ketone (22.8%) and methyl isobutyl ketone (16.4%)]. Aromatics and aliphatics accounted for one each [xylenes (22.8%) in addition to toluene as stated above and *n*-hexane (13.5%)].

Solvents of rare use

It was found that not all of the 7 Group 1 solvents and 40 Group 2 solvents (Table 1) were detected in workroom air. Table 4 shows that 16 solvents were detected in less than 1% of the total cases, and 9 solvents were never detected in the present survey. When classified in terms of Group 1 and 2 solvents, the prevalence was 0% for three solvents (1,2-dichloroethane, 1,2-dichloroethylene and 1,1,2,2-tetrachloroethane) out of 7 Group 1 solvents, and 22 solvents among 40 Group 2 solvents were of less than 1% prevalence (including 6 solvents of 0% prevalence).

Table 4. Solvents of low detection prevalence (<1%)

0%<Prevalence<1%	No detection (Prevalence = 0%)
2-Butanol	Chlorobenzene
Carbon disulfide	Cresol
Carbon tetrachloride	1,2-Dichloroethane (*)
Cellosolve	1,2-Dichloroethylene (*)
Cyclohexanol	Isoamyl alcohol
o-Dichlorobenzene	Methyl *n*-butyl ketone
N, N-Dimethylformamide	Methylcyclohexanol
1, 4-Dioxane	1,1,2,2-Tetrachloroethane (*)
Isoamyl avcetate	1,1,1-Trichloroethane
Isoporpyl acetate	
Methyl acetate	
Methylcellosolve	
Methylcyclohexanone	
n-Pentyl acetate	
n-Propyl acetate	
Trichloroethylene	

(*) Group 1 solvents.

Evaluation in terms of legally defined Administrative Control Classes

Solvent level in each unit workplace was scored for Administrative Control Class 1 (adequate work environment), 2 (intermediary work environment) to 3 (poor work environment necessary to be improved). The rates for Class 1, Class 2 and Class 3 cases with all enterprises in combination were 86.6%, 7.6% and 5.8%. Further classification of scores by enterprise scale (Table 5) showed that the distribution of Class 1, 2 and 3 cases was not uniform depending on the enterprise scale. The proportion of Class 1 cases increased and those of Class 2 and Class 3 decreased as a function of an increase in enterprise scale ($p<0.01$ by Cochran-Armitage's trend test).

The Class 3 cases did not distribute uniformly among the 11 solvent work types, form the lowest (0%) for 4. Hand writing and drawing workplaces and 11. Testing and research workplaces to the highest (20.7%) in 10. Solvent drying workplaces. Statistical evaluation by χ^2 test among types of workplaces with more than 100 cases disclosed that the percentages for workplaces of 3. Printing (14.3%), 5. Surface coating (12.2%) and 10. Solvent drying (20.7%) were significantly ($p<0.01$) higher than the cases for other types of solvent workplaces.

Table 5. Enterprise scale and score by Administrative Control Classes

Enterprise scale	Administrative Control Classes[a]			
	1 (%)	2 (%)	3 (%)	Sum (%)
A	154 (69.1)	39 (17.5)	30 (13.5)	223 (100)
B	539 (82.8)	64 (9.8)	48 (7.4)	651 (100)
C	83 (94.3)	2 (2.3)	3 (3.4)	88 (100)
D	520 (97.2)	9 (1.7)	6 (1.1)	535 (100)
Total	1,296 (86.6)	114 (7.6)	87 (5.8)	1,497 (100)

[a]After Refs. 16 and 17.

Table 6. Organic solvents detected in field surveys in Japan

Authors (yr)	Ref. No.[a]	Solvents identified						Comments
		Over-all	3. Printing	5. Surface coating	6. Adhesive spreaing and 7. Adhesion	8. Degreasing, cleaning and wiping	9. Painting	
Inoue *et al*. (1983[b])	9	(Not reported)	TOL, EA, XYL, EB, MeOH	(Not reported)	(Not reported)	TRI, 1,1,1-T, MeOH, TETRA, TOL	(Not reported)	A whole country samples; see footnote b.
Kumai *et al*. (1983[c])	10	(Not reported)	TOL, IPA, MeOH, XYL , MEK	(Not reported)	TOL, HEX, MEK, MeOH, EA	(Not reported)	TOL, XYL, MEK, EA, IPA	A whole country samples; see footnote c.
Ukai *et al*. (1997)	11	TOL, XYL, MeOH, EA, IPA	TOL, MEK, XYL, IPA, EA	XYL, IPA, TOL, MeOH, EA	TOL, EA, MEK, IPA, MeOH	TOL, DM, MeOH, IPA, Ace	TOL, XYL, MeOH, EA, BA	Cases cited were from Kyoto and its vicinity, Japan.
Yasugi *et al*. (1998)	13	TOL, XYL, EA, MeOH, BA	TOL, IPA, MEK, EA, Cycloh	STY, Ace, TOL, MeOH, DMF	TOL, EA, HEX, MEK, Ace	DM, MeOH, TOL, TETRA, IPA	TOL, EA, BA, MeOH, IBA	Samples from Hiroshima, Japan
Samoto *et al*. (2006)	14	TOL, IPA, MeOH, EA, XYL	TOL, MEK, EA, IPA, MeOH	IPA, MeOH, TOL, XYL, MEK	TOL, EA, MEK, Ace, MeOH	IPA, Ace, TOL, MeOH, EA	TOL, XYL, BA, MeOH, EA	Samples from Kyoto, Japan
The present survey		TOL, IPA, MeOH, Ace, BA	TOL, MEK, EA, IPA, MeOH	TOL, MeOH, IPA, XYL, MEK	TOL, EA, Ace, MEK, MeOH	IPA, Ace, TOL, MeOH, EA	TOL, XYL, BA, EA, BuOH	Samples from Kyoto, Japan

Notes to Table 6
Five solvents of highest prevalence are cited; the order of solvents show the order of prevalence, e.g., the first solvent was of the highest prevalence.
Abbreviations are: BA, *n*-butyl acetate; BuOH, 1-butyl alcohol; DM, dichloromethane; DMF, *N,N*-Dimethylformamide; IBA, isobutyl alcohol; IPA, isopropyl alcohol; MeOH, methyl alcohol; MIBK, methyl isobutyl ketone; STY, styrene; TOL, toluene; XYL, xylenes.
[a]Reference number of publication.
[b]Homogeneous solvent preparations (i.e., thinner for printing and degreasing agents) were analyzed.
[c]Head-space air of heterogenous solvent preparations (i.e., Printing inks and adhesives) were analyzed.

Discussion

The present survey in a whole year period from 2009 to 2010 made it clear that solvent use for degreasing, cleaning and wiping was common irrespective of enterprise size, whereas that for testing and research activities was substantially more common in large-scale enterprises. Printing and painting were most common in medium-scale enterprises. Typical use of solvents was as mixtures (with a median of 5 solvents per sample air), rather than un-mixed single use. Predominant mixture use in industries makes a sharp contrast to the cases in research institutions where solvents were generally used unmixed[15]. Among the regulation-defined Group 1 and 2 solvents (47 types in total), toluene was most common essentially irrespective of types of solvent work (including printing and painting). This was followed by isopropyl alcohol especially in degreasing, cleaning and wiping workplaces. In contrast, 25 solvents (3 solvents in Group 1 and 22 solvents in Group 2) were detected only in limited occasions (i.e., less than 1%). Better working environments (e.g., lower solvent levels in workroom air) in large-scale enterprises are on line with previous findings[12].

Table 6 summarizes results of previous solvent survey by this study group, in comparison with the present findings. It should be noted that the methods of solvent analyses in two early time studies of Inoue *et al*.[9]

Table 7. Solvent components reported in selected latest publications outside of Japan

Authors (year[a])	Ref. No.[b]	Solvents identified	Comments
Moon *et al.* (2001)	28	Toluene (followed by xylenes, hexane, methyl ethyl ketone and acetone) was most common in printing, painting, surface coating and adhesive application workplaces.	Observation in 862 solvent-using worplaces in an industrial complex
Ihrig *et al.* (2001)	26	Toluene, xylenes, butyl acetate, ethylbenzene	Exposure at low concentrations (i.e., <1 ppm each)
Moshe *et al.* (2002)	23	Toluene, xylenes, benzene, methyl ethyl ketone, acetone etc.	Case report on a painter with neuropathy
Ihrig *et al.* (2005)	29	Toluene, xylenes, ethylbenzene, butyl acetate, white spirit	Solvent mixture exposure of painters, printers, etc.
Filley *et al.* (2004)	24	Toluene	A toluene dependency case
Herpin *et al.* (2009)	30	Hexane, toluene, methyl ethyl ketone, acetone	Exposure at adhesive-producing plants
Keski-Säntti *et al.* (2010)	27	Toluene (53%), mineral spirits (47%), butanol (27%), acetone (26%), ethyl acetate (23%), glycol compounds (20%) , etc.	% detection in chronic solvent encephalopahty-inducing solvent preparations; for details, also see the Discussion section.

[a]Year of publication.
[b]Reference number.

and Kumai *et al.*[10] were different from those in other studies[11, 13, 14] (including the present study) in the sense that direct injection (into a gas-chromatograph) of the homogeneous preparations (such as thinners and degreasers) was employed by Inoue *et al.*[9], and headspace air in vessels containing heterogeneous preparations (e.g., inks and paints) was analyzed by Kumai *et al.*[10] for solvent component analyses. The use of solvents as a mixture has been a typical pattern of solvent use in various industries. Botth Ukai *et al.*[11] and Yasugi *et al.*[13] reported that the use of solvents as unmixed accounted for 35% in their surveys, and it was 25% by Samoto *et al.*[14] and 28% in the present survey, indicating that mixture use has been common up to present. When leading (i.e., most frequently detected) solvent was identified (Table 6), it has been toluene in printing, painting and adhesive application workplaces. In case of surface coating, it was toluene in the present survey, but other solvents were listed in the past, e.g., isopropyl alcohol and methyl alcohol by Samoto *et al.*[14] in 2006. Changes in common solvents for 8. Degreasing, cleaning and wiping work should be worthy of noting. Trichloroethylene and 1,1,1-trichloroethane were detected in the early 1980s study by Inoue *et al.*[9], and they were replaced by dichloromethane in late 1990s[11, 13]. Both were further replaced by isopropyl alcohol, acetone and toluene in the present survey, possibly for cleaning and wiping but not for vapor degreasing. The reduction in use of trichloroethylene and 1,1,1-trichloroethane was assumedly in response to environmental concern for prevention of ground water pollution and ozone layer preservation, respectively[21, 22].

Reports from outside of Japan on identification (by chemical names) of solvents in workplaces are rather limited. Literature retrieval gave 7 publications in the past 10 years (Table 7). Some of them were case reports[23–25] or compilations of cases[26, 27], and others were from field surveys[28–30]. Compilation of solvent names reported in these articles clearly suggests that toluene is the leading solvent involved (Table 7).

Among these articles, the reports by Moon *et al.*[28] and Keski-Säntti *et al.*[26] deserve particular attention, as a large number of cases were studied. According to Moon *et al.*[28], analyses of solvents used in 862 workplaces showed that toluene was most frequently detected in painting, printing, adhesive applying and cleaning workplaces, and that the patterns in types of solvents used were generally very similar to Ukai *et al.*[11], Yasugi *et al.*[13] and Samoto *et al.*[14] except that trichloroethylene was in use in cleaning workplaces. Keski-Säntti *et al.*[27] reviewed 129 cases of chronic solvent encephalopathy (CSE) observed during 1995–2007 in Finland. Names of solvents estimated to be causative were filed up in a cumulative manner as more than two solvents were reported for a case. Solvents identified included aromatics (in 78% of cases being the highest, and 53% for toluene), aliphatics, chlorinated hydrocarbons, alcohols, ketones, esters etc. The presence of

CSE symptoms/signs suggests that the cases identified were with heavy solvent exposures. Nevertheless it is possible to understand that toluene was commonly used in solvent workplaces.

The present survey was conducted following legal requirements in Japan, and target solvents were limited to 7 Group 1 Solvents and 40 Group 2 Solvents. This should be taken as the limitation of the present study. As the regulatory list of solvents (Table 1) is rather old although it is still effective, new surrogate solvents[4–6] including water-based solvents[31] or brominated solvents such as 1-bromopropane[32] cannot be taken into account even though they are in use.

Conclusions

As early as in 1980s, toluene has been the most common solvent in over-all solvent workplaces. It is typically so in printing, surface coating, adhesive application, and painting workplaces. This trend was in agreement with use pattern outside of Japan as retrieved through literature survey. In workplaces for degreasing, cleaning and wiping, use of trichloroethylene, and 1,1,1-trichloroethane has ceased to exist, and use of isopropyl alcohol has become common.

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