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SHORT COMMUNICATION

Shift to non-aromatic solvents in solvent workplaces in Japan

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Abstract
Objectives The present analyses were initiated to elucidate changes in types of prevalently used organic solvents in various solvent workplaces in Japan during the past 30 years.
Methods Five publications on industrial solvent use were employed as databases, which were supplemented by solvent workplace surveys in 2010–2012; the surveys were conducted following regulation-stipulated procedures. In practice, data on 500–600 unit workplaces/year were available; the data were classified by types of solvent work according to the regulation.
Results The use of aromatic solvents (typically toluene) has been gradually replaced in a recent few years with methyl alcohol and isopropyl alcohol in surface coating workplaces, whereas toluene has been essentially most prevalent in printing and painting workplaces. In cleaning workplaces, the use of chlorinated hydrocarbon solvents was almost terminated in the years before 2000.
Conclusions The present study may be the first report to note that the prevalence of toluene use has been decreasing in surface coating workplaces to be replaced by alcohols, although toluene has been most prevalent in printing and painting workplaces. In cleaning workplaces, there was a substantial reduction in the use of chlorinated hydrocarbon solvents.

Keywords Alcohols · Aromatic solvents · Organic solvents · Toluene · Work environment

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Introduction

Organic solvents are fundamental industrial chemicals employed in various workplaces. Whereas the toxicities of organic solvents have been gradually understood in industrial communities through regulatory activities [1, 2], types of solvents in use are surveyed only on limited occasions and the annual changes in types of common solvents are seldom reported. Rather exceptionally, this study group has been conducting solvent surveys repeatedly since 1980 [3–7].

The present analysis was initiated to elucidate possible changes in leading solvents in various types of solvent workplaces (printing, painting, etc.). The analysis covered the past 30-year period in Japan, by use of data published in literatures in combination with the results from supplemental surveys conducted in latest 3 years.

Materials and methods

Five publications on survey for solvent use in industrial settings were employed as the databases for the analyses [3–7]. In these reports, the solvent use was surveyed basically in factories in Kyoto prefecture and its vicinities in Japan. The monitoring design was as stipulated by law [1], except for the survey in 1980 when the survey procedures were yet to be defined by regulation. Supplemental surveys were conducted in 2010, 2011 and 2012 in Kyoto prefecture and the vicinities to collect the latest information. Cases of public research institutions were excluded.

In short, the regulatory system considers a hypothetical grid in each solvent workplace, and room air at the 5 or more crosses (together with the site of possible highest concentration) was taken, e.g., into sampling bags. Organic

solvents (7 and 40 types of group 1 and group 2 solvents, respectively, also legally defined [8]) in sampled air are identified by capillary FID-gas chromatography (FID-GC), as previously detailed [5–7]. The detection limit was 1 ppm for all solvents both in the publications and in the supplemental surveys.

In the 1980 study [3, 4], small quantities (e.g., ca. 10 ml) of solvent-containing samples (e.g., paints and inks) were collected in glass bottles with airtight screw caps. A portion of homogeneous solvent liquid (e.g., thinner) or headspace air in the bottle (in case of heterogeneous samples, e.g., paints) was subjected to FID-GC. No materials for surface coating were collected in the 1980 survey, unfortunately [3, 4]. The details of analytical procedures were described elsewhere [3, 4].

Solvent workplaces were grouped after regulatory classification [1, 2, 8], of which five types are selected as major groups, i.e., printing, painting, glue application, surface coating, and cleaning- degreasing- wiping (cleaning in short). Prevalence in the detection of the solvent [%; (the number of cases where the solvent was detected × 100)/ the number of unit workplaces surveyed)] was taken as the indicator of its commonality. Because solvents were often used as mixtures (rather than unmixed) in solvent workplaces [5–7], summation of the prevalence percentage usually exceeded 100 (%).

For statistical evaluation, χ^2 test was applied to detect significant difference in the prevalence of cases.

Results

The three solvents most frequently detected in each type of solvent workplaces were presented in the Table 1, as representative solvents for the survey year.

In 1980, toluene was the most prevalent solvent in printing, painting and glue application workplaces. In contrast, chlorinated hydrocarbon solvents such as trichloroethylene were used typically for vapor degreasing. No data were available for solvents used in surface coating workplaces. Toluene kept the leading position as the most prevalent solvent in printing, painting and glue application workplaces until the first decade in the present century.

Toluene was commonly used also in surface coating workplaces in 2009, but isopropyl alcohol and methyl alcohol took the leading positions in 2010 and later, as to be discussed later. In cleaning workplaces, three chlorinated hydrocarbons of trichloroethylene, 1,1,1-trichloroethane and then dichloromethane were leading solvents in 1980s [3, 4] .The use of trichloroethylene and 1,1,1-trichloroethane were suppressed in reflection of environmental concern [9, 10]. The use of isopropyl alcohol became increasingly common since 2004 survey [6]. It should be noted that two chlorinated hydrocarbon solvents of 1,2-dichloropropane and dichloromethane were recently suspected as the causative chemicals of cholangiocarcinoma, which were detected among proof-printing workers [11]. The 2012 survey in industries, however, did not find any cases of dichloromethane use. It should be added that 1,2-dichloropropane was not in the list of legally defined organic solvents [1, 8] until 2013 and therefore out of the targets of the present survey, unfortunately.

To follow-up further changes, the survey was repeated annually in the latest 3 years (the bottom half in the Table 1). Toluene has been the most prevalent solvent in printing and painting workplaces even in these years, except that toluene prevalence was barely exceeded by that for xylenes in printing workplace in 2012. The first position in glue application was, however, taken by ethyl acetate in 2012 with toluene at the second position.

The changes were further clear in surface coating workplaces. Toluene once took the first position in 2009 but dropped to the third with methyl alcohol and isopropyl alcohol at the first and second positions, respectively. The prevalence of toluene use in surface coating was significantly ($p < 0.01$ by χ^2 test) reduced in 2010–2012 as compared with that in 2009. In the cleaning workplaces, the use of isopropyl alcohol and acetone became more prevalent although toluene maintains the second or third position in the commonality. Since 2004, the prevalence for isopropyl alcohol has been higher ($p < 0.01$ by χ^2 test) than that for toluene.

Discussion

It appears likely that this is the first report to note the reduction in toluene use in surface coating workplaces in recent 3 years. In cleaning workplaces, the use of isopropyl alcohol and probably that of acetone also became more prevalent than that of toluene since 2004. As toluene is a central nervous system depressant [12], the reduction in use may carry substantial industrial health significance. It was not possible in the present analyses to examine if the work environments as a whole have been improved in recent years.

No similar survey on types of solvent use in industries has been conducted in other areas except for Yasugi et al. [13] and Moon et al. [14]. Yasugi et al. [13] observed in Hiroshima prefecture in western Japan that toluene was the most commonly used solvent in printing, painting and glue application workplaces. Moon et al. [14] obtained very similar results of common toluene use in printing, painting and glue application in addition to cleaning workplaces in factories in Busan suburbs in Korea. These articles combined with the report by Ukai et al. [5] may suggest very common use of toluene across various types of solvent

Table 1 Shift in common solvents by type of solvent work

Survey year	Authors and year of publication	Ranking	Type of solvent work										Sum for unit work-places
			Printing	(%)	Painting	(%)	Glue application	(%)	Surface coating	(%)	Cleaning[b]	(%)	
1980	Inoue et al. [3], Kumai et al. [4]	1	Tol	(56)	Tol	(80)	Tol	(51)			TRI	(21)	
		2	IPA	(38)	Xyl	(66)	MeOH	(13)			MC	(14)	
		3	MeOH	(31)	MEK	(28)	Et Ac	(11)			MeOH	(14)	
		No.[c]	Not applicable[d]										
1996	Ukai et al. [5]	1	Tol	(45)	Tol	(61)	Tol	(51)	Xyl	(25)	Tol	(24)	
		2	MEK	(26)	Xyl	(49)	Et Ac	(31)	IPA	(25)	DiChMt	(20)	
		3	Xyl	(26)	MeOH	(39)	MEK	(19)	Tol	(24)	MeOH	(18)	
		No.[c]	68		177		68		101		355		456
2004[a]	Samoto et al. [6]	1	Tol	(61)	Tol	(78)	Tol	(47)	IPA	(51)	IPA	(42)	
		2	MEK	(55)	Xyl	(76)	Et Ac	(42)	MeOH	(43)	Acet	(34)	
		3	Et Ac	(47)	*n*-BuAc	(57)	MEK	(33)	Tol	(36)	Tol	(25)	
		No.[c]	131		161		55		106		314		420
2009[a]	Nagasawa et al. [7]	1	Tol	(61)	Tol	(82)	Tol	(49)	Tol	(82)	IPA	(49)	
		2	MEK	(55)	Xyl	(77)	Et Ac	(45)	MeOH	(76)	Acet	(34)	
		3	Et Ac	(47)	*n*-BuAc	(65)	Acet	(32)	IPA	(64)	Tol	(31)	
		No.[c]	154		182		111		115		452		567
2010[a]	The present study	1	Tol	(63)	Tol	(79)	Tol	(51)	MeOH	(51)	IPA	(50)	
		2	Et Ac	(60)	Xyl	(73)	Et Ac	(42)	IPA	(45)	Acet	(32)	
		3	MEK	(58)	*n*-BuAc	(63)	MeOH	(42)	Tol	(45)	Tol	(27)	
		No.[c]	153		205		117		112		527		639
2011[a]	The present study	1	Tol	(63)	Tol	(79)	Tol	(48)	MeOH	(49)	IPA	(52)	
		2	IPA	(56)	Xyl	(77)	Et Ac	(38)	IPA	(43)	Acet	(29)	
		3	Et Ac	(56)	*n*-BuAc	(55)	MeOH	(27)	Tol	(38)	Tol	(29)	
		No.[c]	143		184		148		136		518		654
2012[a]	The present study	1	Tol	(59)	Xyl	(79)	Et Ac	(49)	MeOH	(49)	IPA	(50)	
		2	MEK	(53)	Tol	(75)	Tol	(43)	IPA	(47)	Tol	(35)	
		3	IPA	(50)	*n*-BuAc	(66)	Xyl	(26)	Tol	(44)	Acet	(26)	
		No.[c]	115		159		87		116		346		462

Acet acetone, *DiChMt* dichloromethane, *Et Ac* ethyl acetate, *IPA* isopropyl alcohol, *MC* methylchloroform (1,1,1-trichloroethane, *MEK* methyl ethyl ketone, *MeOH* methanol, *n-BuAc* *n*-butyl acetate, *Tol* toluene, *TRI* trichloroethylene, *Xyl* xylenes

[a] From 1 April of the year to 31 March of the next year

[b] Cleaning, degreasing and wiping

[c] Number of unit workplaces

[d] For reasons, see "Materials and methods" section

workplaces both in Japan and in Korea in the years around 2000. However, it is not known whether shift to non-aromatic solvents (especially to alcohols) is currently taking place also in these places.

In conclusion, there has been continuous trend of less use of aromatic solvents (typically toluene) especially in surface coating workplaces. In cleaning workplace, isopropyl alcohol and possibly acetone are more prevalently used than toluene.

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Conflict of interest The authors declare that they have no conflicts of interest.

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