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Indoor Emission Characteristics of Liquid Household Products using Purge - and - Trap Method

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Abstract

Since the emissions composition from the household products have potentially been associated with health risks for building occupants, the chemical composition emitted from the products should be surveyed. The current study identified the emission composition for 42 liquid household products, using a purge-and-trap method. This evaluation was done by classifying the household products into five product classes (deodorizers, household cleaners, color removers, pesticides, and polishes). Nineteen compounds were chosen on the basis of selection criteria. The quality control program for purge-and-trap and analytical systems included tests of laboratory blank Tenax traps and blank water samples, and the determination of calibration equation, measurement precision, method detection limit (MDL), and recovery. The number of chemicals varied according to the product categories, ranging from 4 for the product category of bleaches to 12 for the product categories of air fresheners and nail color removers. For all product categories, the emission composition and concentrations varied broadly according to product. It is noteworthy that most household products emit limonene: 19 of 25 cleaning products; 5 of 6 deodorizers; 1 of 3 pesticides; 3 of 3 color removers; and 4 of 5 polishes. It was suggested that the use of household products sold in Korea could elevate the formation of secondary toxic pollutants in indoor environments, by the reaction of limonene with ozone, which entered indoor environments or might be generated by indoor sources such as electronic air cleaning devices and copying machines.

Keywords: Color remover, Composition, Deodorizer, Household cleaner, Pesticide, Polisher

1. Introduction

While building materials have received a great concern associated with indoor air quality,[1,2] household products have received a less concern. Certain studies have suggested these household products as sources of indoor air pollutants (IAPs),[3,4] although they perceived benefits of human life such as promotion of hygiene and aesthetics.[5-7] In fact, these IAPs have been prevalent in indoor air of households or public buildings due to the variety of indoor sources including the uses of several household products.[8-10] Many IAPs have been shown to cause a group of symptoms including sleepiness, irritability, inability to concentrate, and other health hazards.[11,12] A major cause of these health effects for building occupants is the inhalation exposure to consumer-product constituents and the secondary pollutants produced by the reaction of unsaturated organic constituents with oxidants.[12-15] The inhalation exposure of indoor air pollutants associated with the use of the consumer products include the inhalation of nonvolatile components as well as volatile components.[4] However, present study focused on the volatile components emitted directly from liquid consumer products.

Since the emissions composition from the household products has potentially been associated with health risks for building occupants, the chemical composition emitted from the products should be surveyed.[16] Nevertheless, very limited information is available about the emissions composition for the household products available in many countries. Chemical components and their proportions emitted from consumer products are suspected to be different among manufacturers in different countries. Consequently, the current study evaluated the chemical composition for liquid consumer products sold in Korea, using a semiquantitative purge-and-trap method. A compact group of 19 target compounds was selected for this study, based on their detection frequencies in indoor air of households or public buildings,[8,10] their reactivity with oxidants for the formation of potentially harmful secondary pollutants,[12-15] or their own toxicity[17].

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2. Experimental Methods

Forty-two liquid household products were selected for this study and organized into 16 product categories which were further organized into 5 product classes (deodorizers, household cleaners, color removers, pesticides, and polishes). The products were selected on the basis of sales figures, i.e. the number of items sold during the previous year. The sales figures were obtained from the sales personnel of the three largest supermarket companies in Korea. All household products were purchased from three supermarket companies less than one year after being manufactured.

For the 19 target compounds screened by the selection criteria, a purge-and-trap method was used to identify their proportions in the purged-gas phase by applying the method employed by Sack et al.,[3] using a GC (Varian 3400CX) equipped with a flame ionization detector (FID) and a capillary column (Supelco Co. SPB-5). The selection criteria included their detection frequencies in the headspace phase and their reactivity with oxidants for the formation of potentially harmful secondary pollutants or their own toxic effects. A solution of each consumer product was prepared by adding 100 µL of raw material to 1 to 5 mL of high-purity methanol (Aldrich, HPLC grade), which is dependent on the formation of bubble in the purge vessel. Then this solution was added to 5 mL of purified water in a 25-mL purge vessel. Nitrogen was bubbled for 30 min at room temperature (19 to 23°C) through the vessel to allow for evolution of volatile pollutants, if any, from each of the materials, which were trapped on a Tenax TA trap. The adsorbent trap was thermally desorbed at 250°C for 10 min, and the target compounds cryofocussed at -120°C on a cryo trap (15.2-cm-long, 0.32-cm-o.d. tube packed with glass beads). The cold trap was rapidly heated to 250°C, then the contents were flushed into the Cryofocusing Module (CM) of the TDS and cooled to -120°C to refocus the target compounds. The CM was then heated to 225°C and flushed to transfer the target compounds to the GC. The initial oven temperature was set at 35°C for five min and ramped at 4°C/min to 200°C for five min. The mass concentrations of each analyte were reported to present the relative chemical proportions in purged gaseous phase.

The quality control program for purge-and-trap GC/FID system included tests of laboratory blank Tenax traps and blank water samples, and the determination of calibration equation, measurement precision, method detection limit (MDL), and recovery. Six-point calibration measurements were made for the target VOC. To check the quantitative response of the analytical system, a known standard was directly injected into the GC. The precision and MDL (United States Environmental Protection Agency Method) of the target VOC were determined by seven repeated analyses of one of the calibration standard solutions. One mL of high-purity methanol spiked with known amounts of the target analytes was added to 5 mL of purified water in the purge vessel to establish recovery for the determination of the target compounds for purge-and trap method.

3. Results and Discussion

3.1. Quality Control

Table 1 exhibits the results of quality control program: precision, MDL, and recovery of the purge-and-trap GC/FID method. The precision of the system was characterized by the relative standard deviation of the measurements. The precisions of the analytes fell within the range of 0-20% recommended in EPA Method 624. The MDL of target compounds ranged from 0.5 to 24.5 ng/µL for 100 µL of raw material. The recoveries of all target compounds fell within the range of 80-110% except for 2- butoxy ethanol and 1,4-dioxane. The recoveries for 2-butoxy ethanol and 1,4-dioxane were 72% and 53%, respectively, which are miscible in water and would not be purged with high efficiency. Similarly, Sack et al.[3] determined the lowest recovery of 46% for 1,4-dioxane from their purge-and-trap analysis of household products sold in America. Concentration values reported in the database were not corrected for recovery.

Table 1. Precision, MDL, and recovery of the purge-and-trap GC/FID method determined for the determination of 19 target VOCs

Analyte	Precisions (%)	MDL (ng/µL)	Recovery (%)
Acetone	12.5	12.7	88
Benzene	12.1	15.6	99
2-Butoxy ethanol	12.0	4.3	72
Chlorobenzene	6.1	1.4	88
Chloroform	12.2	1.8	91
Decane	10.7	7.1	96
1,4-Dioxane	8.0	1.5	53
Ethanol	12.5	10.4	83
Ethylbenzene	12.4	0.5	108
Hexane	10.0	4.5	95
Limonene	12.4	6.7	88
Phenol	6.3	5.7	90
1-Propanol	11.2	24.5	82
Toluene	12.1	1.5	103
PCE (Tetrachloroethylene)	9.9	1.2	94
TCE (Trichloroethylene)	9.0	1.0	98
m,p-Xylene	10.0	1.9	102
o-Xylene	9.1	4.2	94

The experimental system performance was also evaluated for system contamination and the response of GC system. At the beginning of the day, the laboratory blank Tenax traps and blank water samples were analyzed to check for any contamination during the sampling and analytical procedures. However, no trap or water contamination was identified. In one day when the quantitative response of FID differed by more than ±20% from that predicted by a specified calibration equation, a new calibration equation was determined.

3.2. Emission Composition and Concentrations

Table 2 exhibits the number of target compounds measured in headspace phases of 42 household products. The number of chemicals varied according to the product categories, ranging from 4 for the product category of bleaches to 12 for the product categories of air fresheners and nail color removers. Although the number of products analyzed was different between the product categories, the difference is not significant with a range of one to five for the number of products analyzed. Moreover, the number was two or three for most cases. As such, the number of products analyzed would not significantly influence the number of target compounds in each product category. Most of compounds measured in the present study were also found in furniture wax sold in the U.S. from headspace tests reported by Wallace et al.[18].

Table 3 shows the concentrations of target compounds identified in purged gas of 25 cleaning products. The emission composition and concentrations varied broadly according to product. This difference might be attributed to the combined effects of component volatility and the matrix of each product. The emissions composition is a function of the volatility of the components and their concentrations in the liquid phase.[19] Among four compounds, ethanol was the highest compound emitted from bleaches. For dishwashing detergents, three compounds which have relatively high emission concentrations are ethanol,

Table 2. Number of compounds identified in headspace phases of 42 household products

Product class	Product category	Number of products Analyzed	Number of compounds identified
Cleaning products	Bleaches	1	4
	Dishwashing detergents	3	7
	Disinfectants	5	10
	Dry cleaning products	2	6
	Fabric softeners	3	6
	General purpose cleaners	3	9
	Glass cleaners	2	6
	Laundry detergents	2	7
	Laundry stain removers	2	7
	Oven cleaners	2	9
Deodorizers	Air fresheners	4	12
	Fabric deodorizers	2	5
Pesticides	Liquid pesticides	3	8
Color removers	Nail color removers	3	12
Polishes	Furniture polishes	3	9
	Nail polishes	2	7

Table 3. Concentrations of selected compounds in purged gas of 25 cleaning products

Product class	Product category	Product	Compounds	Conc (μg/μL of raw materials)
Cleaning products	Bleaches	BLG	Ethanol	6.2
			Limonene	0.88
			PCE	0.45
			Toluene	0.11
	Dishwashing detergents	DCG	Decane	0.02
			Ethanol	1.51
			Limonene	1.14
			PCE	0.44
			Phenol	0.04
			1-Propanol	2.07
			o-Xylene	0.04
		DDL	Ethanol	1.2
			Limonene	1.61
			PCE	0.34
			Phenol	0.04
			1-Propanol	1.77
		DJP	Ethanol	1.8
			Limonene	4.06
			1-Propanol	1.65
	Disinfectants	DHS	Ethanol	1.07
		DNB	2-Butoxy ethanol	0.01
			Ethanol	3.97
			Limonene	0.09
			PCE	0.44
			Phenol	0.04
			m,p-Xylene	0.005

Disinfectants	DOS	Chlorobenzene	0.003
		Decane	0.05
		Ethanol	2.57
		Limonene	0.13
		PCE	0.04
	DSS	2-Butoxy ethanol	0.009
		Ethanol	1.51
		PCE	0.035
		1-Propanol	1.36
		Limonene	0.03
	DPZ	Acetone	2.34
		Decane	0.02
		Ethanol	1.26
		Limonene	1.09
		PCE	0.46
		Phenol	0.02
		1-Propanol	14.09
Dry cleaning products	DHD	Limonene	1.65
		PCE	0.22
		Phenol	0.26
	DHJ	Chlorobenzene	0.05
		PCE	0.34
		Phenol	0.16
		1-Propanol	0.72
		Toluene	0.08
Fabric softeners	FF	Ethanol	0.92
		Limonene	0.11
		PCE	0.23
	FP	Ethanol	1.67
		Hexane	1.02
		Limonene	0.05
	FS	Acetone	2.18
		Ethanol	6.46
		Limonene	0.09
		Phenol	0.01
General purpose cleaners	AD	Acetone	2.47
		Decane	0.07
		Toluene	0.03
	AF	Benzene	0.001
		1,4-Dioxane	0.009
		Ethanol	1.28
		Limonene	0.04
		Toluene	0.007
	AM	Chloroform	9.26
		PCE	0.22
Glass cleaners	GH	Acetone	8.42
		Limonene	0.02
		PCE	0.25
		1-Propanol	1.58
	GW	2-Butoxy ethanol	0.02
		Ethanol	0.41
		Limonene	0.07
Laundry detergents	LB	Chlorobenzene	0.03
		Ethanol	10.90
		PCE	0.48
	LT	Acetone	0.02
		Benzene	0.01
		Chlorobenzene	0.06

			Ethanol	1.67
			Limonene	0.13
			PCE	0.68
			Phenol	0.04
	Laundry stain removers	LO	Acetone	4.86
			Chlorobenzene	0.04
			Ethanol	1.14
			PCE	0.58
		LS	Acetone	2.11
			Decane	0.04
			Limonene	0.02
			PCE	0.04
			1-Propanol	1.90
	Oven cleaners	OG	Acetone	8.39
			Limonene	2.61
			PCE	0.29
		OO	2-Butoxy ethanol	0.01
			Decane	0.03
			Ethanol	2.83
			Ethylbenzene	0.007
			Limonene	0.41
			PCE	0.15
			Phenol	0.1
			o-Xylene	0.004

limonene, and 1-propanol. For most disinfectants, ethanol exhibited the highest emission concentration. For dry cleaning products, PCE and phenol were identified in two products. Limonene and ethanol were identified in all three fabric softeners. The chemical composition of general purpose cleaners were mostly different among the three products. For glass cleaners, acetone was measured at a substantially high emission concentration (8.42 µg/µL of raw materials) as compared to other compounds. Ethanol was emitted at the highest concentration in two laundry detergents. For two laundry stain removers, acetone was emitted at the highest concentration. Acetone was also emitted at the highest concentration in one oven cleaner (OG), but it was not identified in other oven cleaner (OO).

The emission concentrations of target compounds identified in purged gas of six deodorizers are presented in Table 4. The emission composition and concentrations varied broadly according to the products. Limonene showed the highest emission concentration in three (AD, AG, and AL) of four air fresheners. Phenol was found in one air freshener (AW). Limonene was found in two fabric deodorizers, but its emission concentrations were substantially lower than three air fresheners (AD, AG, and AL). Meanwhile, ethanol was emitted at the highest concentra-

Table 4. Concentrations of selected compounds in purged gas of six deodorizers

Product class	Product category	Product	Compounds	Conc (µg/µL of raw materials)
Deodorizers	Air fresheners	AD	Acetone	3.49
			Decane	0.67
			Ethanol	1.57
			Hexane	2.16
			Limonene	5.42
		AG	Ethanol	0.05
			Ethylbenzene	0.002
			Limonene	0.37
			Toluene	0.006
			m,p-Xylene	0.002
		AL	Benzene	0.003
			Decane	1.33
			Ethanol	0.98
			Limonene	2.69
			1-Propanol	0.67
		AW	Decane	0.85
			Ethanol	1.41

			Hexane	2.07
			Phenol	2.16
			1,1,1-Trichloroethane	1.02
	Fabric deodorizers	FF	2-Butoxy ethanol	0.03
			Ethanol	7.14
			Limonene	0.04
			PCE	0.29
		FS	Ethanol	4.4
			Limonene	0.06
			PCE	0.37
			Toluene	0.08

tion in both fabric deodorizers.

Table 5 exhibits the emission concentrations of target compounds measured in purged gas of three pesticides. Similar to cleaning products and deodorizers, the emission composition and concentrations varied according to the products. However, decane, PCE, phenol, and o-xylene were found in all three liquid pesticides. Limonene and toluene were identified in only one product.

The emission concentrations of selected compounds detected in purged gas of three color removers are presented in Table 6. Similar to pesticides, the emission composition and concentrations varied according to the products. However, acetone, limonene, and PCE were found in all three liquid pesticides. Acetone showed the highest emission concentration in the three liquid pesticides. 1,4-dioxane was detected in one nail color remover (NC), although its emission concentration is not high compared with other compounds.

Table 7 shows the emission concentrations of selected compounds detected in purged gas of five polishes. Similar to color removers, the emission composition and concentrations varied according to the products. However, decane and limonene were found in all three furniture polishes. Limonene exhibited the highest emission concentration in the three furniture polishes. 2-Butoxy ethanol was found in two nail polishes. This compound has been employed in variety household products[12,19,20]: all purpose cleaners, glass and surface cleaners, antibacterial glass and surface cleaners; lemon fresh and antibacterial spray; nail removers; surface coating products; and caulking and sealants. The use of 2-butoxy ethanol along with 2-methoxyethanol and 2-ethoxyethanol have raised health concerns.[21)]

It is noteworthy that most household products emit limonene: 19 of 25 cleaning products (Table 3); 5 of 6 deodorizers (Table 4); 1 of 3 pesticides (Table 5); 3 of 3 color removers (Table 6); and 4 of 5 polishes (Table 7). Similarly, several studies[19,22-26)] conducted in other countries identified limonene in various household products. The household cleaning products which are reported to emit limonene include furniture polish, antibacterial glass and surface cleaner, disinfectant, floor shine cleaner for no-wax and regular floors, general purpose cleaner, glass cleaner, and lemon freshener and antibacterial spray.[19)] Limonene and other terpenes are added to household products due to their favorable odor and solvent properties.[4)] Terpenes receive a great concern, because they potentially react with ozone to form secondary pollutants such as formaldehyde, hydroxyl radicals, hydrogen peroxide, nitrogen oxides, and organic aerosol.[13-15,23-26)] Consequently, the present finding suggests that the use of house-

Table 5. Concentrations of selected compounds in purged gas of three pesticides

Product class	Product category	Product	Compounds	Conc (µg/µL of raw materials)
Pesticides	Liquid pesticides	PA	Decane	0.33
			Ethanol	0.97
			PCE	0.26
			Phenol	0.57
			1-Propanol	0.85
			o-Xylene	0.22
		PO	Decane	0.47
			Ethanol	1.01
			Limonene	4.04
			PCE	0.46
			Phenol	1.05
			1-Propanol	1.09
			Toluene	0.02
			o-Xylene	0.07
		PU	Decane	4.31
			PCE	0.19
			Phenol	1.21
			o-Xylene	1.04

Table 6. Concentrations of selected compounds in purged gas of three color removers

Product class	Product category	Product	Compounds	Conc (μg/μL of raw materials)
Color removers	Nail color removers	NC	Acetone	2.23
			Chlorobenzene	0.02
			1,4-Dioxane	0.14
			Ethanol	0.7
			Hexane	1.22
			Limonene	0.24
			PCE	0.06
			Phenol	0.05
		NE	Acetone	5.92
			Limonene	0.65
			PCE	0.05
			Phenol	0.37
			1-Propanol	0.52
		NR	Acetone	5.36
			2-Butoxy ethanol	0.02
			1,4-Dioxane	0.02
			Ethanol	1.44
			Limonene	0.14
			PCE	0.02
			1-Propanol	0.5
			TCE	1.09
			o-Xylene	0.02

Table 7. Concentrations of selected compounds in purged gas of five polishes

Product class	Product category	Product	Compounds	Conc (μg/μL of raw materials)
Polishes	Furniture polishes	FO	Decane	0.03
			Limonene	2.17
		PP	Decane	0.03
			Ethylbenzene	0.003
			Limonene	0.7
			PCE	0.22
			Phenol	0.11
			1-Propanol	0.74
		PW	Decane	0.02
			Ethanol	7.69
			Hexane	2.16
			Limonene	12.46
			PCE	0.28
			Phenol	0.14
			1-Propanol	1.31
			Toluene	0.05
	Nail polishes	NE	Acetone	5.53
			2-Butoxy ethanol	0.08
			Ethanol	4.61
			Ethylbenzene	0.13
		NV	2-Butoxy ethanol	0.06
			Ethylbenzene	0.01
			Limonene	0.04
			Phenol	0.02
			o-Xylene	0.02

hold products sold in Korea can elevate the secondary toxic pollutants in indoor environments, by the reaction of limonene with ozone, which enters indoor environments or may be generated by indoor sources such as electronic air cleaning devices and copying machines.

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