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Characterization of emissions composition for selected household products available in Korea

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Abstract

The present study investigated the emission composition for 59 household products currently sold in Korea, using a headspace analysis. The chemical composition and concentrations of total volatile organic compounds (VOCs) broadly varied along with products, even within the same product category. Up to 1–17 organic compounds were detected in the headspace gas phase of any one of the products. The chemical composition of certain household products determined in the current study was different from that of other studies from other countries. Between 4 and 37 compounds were detected in the headspace gas phase of each product class. Several compounds were identified in more than one product class. Of the 59 household products analyzed, 58 emitted one or more of the 72 compounds at chromatographic peak areas above 10^4. There were 11 analytes which occurred with a frequency of more than 10%: limonene (44.2%), ethanol (30.5%), acetone (18.6%), α-pinene (18.6%), *o*,*m*,*p*-xylenes (18.6%), decane (17.0%), toluene (17.0%), β-myrcene (11.9%), ammonia (10.2%), ethylbenzene (10.2%), and hexane (10.2%).

Keywords: VOCs; Headspace analysis; Household product category; Product class

1. Introduction

Even though household products provide substantial benefits to human life, such as the promotion of hygiene and aesthetics, the emissions composition from such products has been a subject of concern in recent years because many of these compounds are potentially associated with health risks for building occupants. Several studies have implicated these consumer products as sources of indoor air pollutants [1–6]. These compounds have been shown to cause symptoms similar to those characterized as Sick Building Syndrome; this is a group of symptoms that includes sleepiness, irritation, inability to concentrate, and other health hazards [7–11]. A major cause of health-related problems for building occupants is the inhalation of consumer-product constituents.

Furthermore, secondary toxic pollutants are formed by the reaction of unsaturated organic constituents with oxidants such as ozone, hydroxyl radicals, and nitrogen oxides [12–14]. For example, terpene, a major constituent of household products such as cleaning products and air fresheners [15,16], reacts with ozone thus leading to the formation of formaldehyde [13,14]. However, the present study focuses only on primary VOCs emitted from household products.

Although direct indoor-air monitoring data can be employed to properly estimate the inhalation exposure of building occupants, the characterization of emissions composition for household products can provide valuable information for the semiquantitative estimation of inhalation exposure and for the selection of safer consumer products. A headspace measurement method has been employed for a semiquantitative determination of volatile components emitted from the consumer products [17]. Meanwhile, a means for carrying out a quantitative analysis in estimating VOCs from the emission of consumer products are small environmental test chambers [18–20]. This kind of survey has been conducted by several research groups, mainly in Western Europe and the USA [1,17–20]. However, information about chemical components emitted from household products employed in many other countries seems to be still

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relatively scant and insufficient. The emissions composition for household products is likely different among manufacturers in different countries. Consequently, the present study investigated the emissions composition for household products currently sold in Korea by using a headspace analysis [21], which has previously been employed for the determination of VOCs in various consumer products.

2. Experimental

2.1. Household products for tests

Consumer products were organized into 8 product classes which were further organized into 21 product categories (Table 1). Fifty-nine household products were selected for the present study. The products were selected on the basis of sales figures for the previous year. The sales figures were obtained from sales persons from three of the largest supermarkets in Korea. All six paints (three oil and three water-based paints) were received from the manufacturers between 3 and 6 months after being manufactured, and other household products were purchased from one of the three supermarkets less than 1 year after being manufactured.

2.2. Determining the emissions composition

The emissions composition in headspace gaseous samples was tentatively determined using a gas chromatograph (HP 5890II) and mass spectrometer (HP MSD5973) (GC/MS) system. Static headspace analysis was conducted to determine the VOCs emitted from household products. This was done by applying the method employed by the USEPA [21].

Table 1
Number of products surveyed in this study according to product class and category

Product class	Product category	Number of products
Cleaning products	Bleach	2
	Dishwashing detergent	4
	Disinfectants	5
	Dry cleaning	2
	Fabric softeners	4
	General purpose cleaners	3
	Glass cleaners	2
	Laundry detergents	3
	Laundry stain removers	2
	Oven cleaners	2
Deodorizers	Air fresheners	4
	Fabric deodorizers	2
Glues	Floor glues	2
	Wallpaper glues	2
Nail color removers	Nail color removers	3
Paints	Oil paints	3
	Water-based paints	3
Pesticides	Liquid pesticides	3
Polishes	Furniture polishes	3
	Nail polishes	2
Sealants	Household sealants	3

For the headspace tests, 8 ml of a household product, except for paints and glues, were placed in a 40-ml clean glass bottle fitted with a Teflon-septum, top screw-cap. For paints and glues, sample materials occupied just roughly 20% of the total volume of the bottle because of difficulties in measuring due to their high viscosity levels. The glass bottles were placed in a water bath at 60 °C for 90 min to allow for the VOCs if any to pass to the gas phase from each of the materials.

For this experiment, 1 ml of gaseous samples was drawn into a 10-ml pressure lock syringe, and transferred to the GC/MS system. A 30-m long fused silica capillary column (internal diameter 0.32 mm; film thickness 1 μm) (Agilent Technologies, HP-5) was used to separate the target analytes. The GC oven was programmed initially at 40 °C for 5 min, and the temperature increased at a rate of 5 °C min^{-1} up to 250 °C, which was held for 5 min.

Compounds were identified using a Wiley mass spectral library. A spectral search quality of 70% was employed for the criterion of the compound selections. Neither the compounds, with a spectral search quality of less than 70%, nor those which were detected in insignificant amounts (chromatographic peak area $<10^4$), were included in the current paper. The chemical name of the first match on the list suggested by the MS library was used and there might be possible that the peak represents other similar structures. Certain overlapping or unresolved peaks may hinder the accurate characterization of chemical composition for the consumer products. The concentration levels of "total VOCs" were calculated by using integrated chromatographic peak areas of analytes (between *n*-hexane and *n*-hexadecane), which were converted to toluene-equivalent concentration for analytes in the headspace gas phase of each household product. Since *m*-xylene and *p*-xylene were co-eluted, the added concentrations of the two compounds were reported.

3. Results and discussion

3.1. Chemical composition according to product

The chemical composition and concentrations of total VOCs, determined from the headspace gas phase according to product, are presented in Table 2 . A total of 59 household products currently sold in Korea were analyzed for VOCs, as potential sources of indoor-air pollution. The chemical composition and concentrations of total VOCs broadly varied with products. One product (ULD) in the category of laundry detergents did not emit any VOCs. Except for this product, up to 1–17 organic compounds could be detected in the headspace gas phase of any one of the other products. Oil paints exhibited an upper range for both the number of chemicals detected and the concentrations of total VOCs. Moreover, the chemical composition and concentrations of total VOCs were substantially different between products even within the same product category. For oil paints, the number of chemical compositions was between 8 and 17, and the concentrations of total VOCs ranged between 3840 and 13,107 ng/ml. As expected, both the number of chem-

Table 2
Chemical composition and concentrations of total VOCs determined from headspace phase according to product

Product class	Product category	Product	Analytes	Concentration (ng/ml)
Cleaning products (37)	Bleach	LLG	Acetone, chloroform, 1,8-cineole, limonene, *cis*-limonene oxide, *trans*-limonene oxide, α-pinene, β-pinene	1,335
		OC	Acetone, chloroform, limonene	3,635
	Dishwashing detergent	CG	Ethanol, limonene	2,412
		DA	Limonene, 1-propanol	1,653
		DL	1,4-Dioxane, ethanol, ethyl acetate, limonene, *β*-myrcene, 3-pentanol, α-pinene	1,886
		JP	Ethanol, 1-hexadecanol, limonene, *β*-myrcene, α-pinene, 1-tetradecane	2,119
	Disinfectants	HS	*iso*-Amyl acetate, ethanol, hexane, limonene, 3-methyl pentane, undecane	1,010
		NB	Camphene, decane, ethanol, hexane, limonene, octane, α-pinene, β-pinene, toluene	1,001
		OS	Ammonia, hexane, limonene, decane	6,967
		PZ	Ethanol	4,252
		SC	Chloroform, 1,8-cineole, *iso*-cineole, ethanol	1,063
	Dry cleaning	HDC	Limonene	8,489
		HDCJ	Ammonia, δ-3-carene, limonene, β-myrcene, octyl aldehyde, α-pinene, sabinene	4,159
Cleaning products	Fabric softeners	DO	Ethanol, limonene, α-pinene	1,416
		FS	2-Butanol, limonene	1,276
		PZO	Ethanol	1,902
		SH	Methoxy ethane, 2-methoxy propane, limonene	1,800
	General purpose cleaners	FP	Decane, limonene, β-myrcene, octane, α-pinene, β-terpinene	1,169
		GN	1,8-Cineole, *iso*-cineole	878
		MI	Ammonia, chloroform	4,167
	Glass cleaners	HS	2-Butoxy ethanol, ethanol, limonene	4,026
		WI	1,8-Cineole, ethanol, limonene	1,989
	Laundry detergents	BI	Acetone, benzene, ethanol	956
		TE	Limonene	35
		ULD	Not detected	
	Laundry stain removers	OCO	Ammonia, decane	1,544
		SNW	Acetone, 3,7-dimethyl-3-octanol	1,156
	Oven cleaners	GZ	Camphene, 1,8-cineole, *iso*-cineole, ethanol, limonene, β-myrcene, α-pinene, sabinene, α-terpinolene	661
		OC	Ammonia, ethanol	1,824
Deodorizers (7)	Air fresheners	AL	Decane, limonene	1,354
		AW	Acetaldehyde, decane, dodecane, ethyl acetate, nonadecane, undecane	5,132
		DFA	Camphene, ethanol, α-pinene	1,356
		GR	Hexane, 2-methyl pentane	1,354
	Fabric deodorizers	FB	Ammonia, ethanol	1,069
		SP	1-Chloro-2-methyl benzene, ethanol, limonene, β-myrcene, α-pinene, β-pinene, γ-terpinene	1,987
Glues (8)	Floor glues	FA	Acetone	1,278
		OD	Ethylbenzene, toluene, *o*-xylene, *m*,*p*-xylene	2,611
	Wallpaper glues	OB1	Ethylbenzene, ethyl acetate, methyl acetate, toluene, *m*,*p*-xylene	1,932
		OB5	2,3-Butanedione, ethyl acetate, methyl acetate	1,680
Nail color removers (4)	Nail color removers	ET	Acetone	5,496
		NC	Acetone, cyclohexane	3,698
		NR	Acetone, butyl acetate, 2,2,4-trimethyl-1,3-dioxolane	1,335
Paints (21)	Oil paints	AS	Acetone, ethylbenzene, ethyl cyclohexane, 4-methyl-2-pentanone, toluene, 2,2,4-trimethyl pentane, *o*-xylene, *m*,*p*-xylene	3,840
		EB	Butyl benzene, butyl cyclohexane, decane, 2,6-dimethyl-heptane, dodecane, ethylbenzene, ethyl cyclohexane, 1-ethyl-3-methyl benzene, 2-methyl-nonane, 4-methyl-2-pentanone, 1-methyl propyl benzene, nonane, propyl cyclohexane, toluene, 2,2,4-trimethyl pentane, *o*-xylene, *m*,*p*-xylene	13,107

Table 2 (*Continued*)

Product class	Product category	Product	Analytes	Concentration (ng/ml)
		SH	Acetone, ethylbenzene, ethyl cyclohexane, 4-methyl-2-pentanone, propyl cyclopentane, toluene, *o*-xylene, *m*,*p*-xylene	7,916
	Water-based	AWT	Octane, *iso*-octane, 2,2,4-trimethyl pentane	1,834
		EF	2,2,4-Trimethyl pentane	1,253
		ST	Ethylbenzene, *m*,*p*-xylene	1,523
Pesticides (7)	Liquid pesticides	OS	Dodecane, limonene, 2-methyl-butane, tetradecane, tridecane, undecane	5,098
		RA	Decane, dodecane, 2-methyl-butane, tetradecane, tridecane	2,850
		RU	Decane	13
Polishes (12)	Furniture polishes	OG	Acetone, 2,7-dimethyl-undecane, limonene, β-myrcene, α-pinene	1,354
		PR	Hexane, limonene	1,252
		WP	Decane, limonene	1,167
	Nail polishes	AVO	Benzene, ethyl acetate, limonene, 2-propanol, toluene	5,729
		ET	Ethanol, toluene	1,523
Sealants (5)	Household sealants	DC	Methyl cyclobutane, methyl cyclopentane, toluene	1,627
		KR	Toluene	2,800
		SN	Hexane, methyl ethyl ketone	1,253

Values in parenthesis represent the number of compounds identified in the respective product classes; the chemical name of the first match on the list suggested by the MS library was used and there might be possible that the peak represents other similar structures.

icals detected and the concentrations of total VOCs were much higher for oil paints than for water-based paints.

The chemical composition of certain household products, determined in the current study, was different from that of other studies conducted in other countries. For example, general purpose cleaners analyzed in the present study emitted six compounds (decane, limonene, β-myrcene, octane, α-pinene, and β-terpinene), whereas in Zhu et al.'s Canadian headspace study [16] five compounds (2-butoxy ethanol, Camphene, limonene, β-myrcene, and β-pinene) were emitted. Similarly, the present study found wallpaper glues to emit five compounds (ethylbenzene, ethyl acetate, methyl acetate, toluene, and *m*,*p*-xylene), whereas Wallace et al. [22] reported that in headspace analyses among 17 target chemicals only just one compound (1,1,1-trichloroethane) was found in wallpaper glues sold in America. Although two compounds (ethylbenzene and *m*,*p*-xylene), which were found in wallpaper glues analyzed in the present study, belonged to the target compounds in Wallace et al.'s study [22], these compounds were not found in this previous study.

The chemical composition and concentrations of total VOCs emitted from each product did not equal those in the raw products. The chemical composition and concentrations of total VOCs in the headspace gas phase are a function of the volatility of the components and their concentrations in the liquid phase [23]. However, headspace tests are useful as a screening tool in selecting target compounds for further study of emissions from household products.

3.2. Chemical composition according to product classes

Table 2 also exhibits the chemical composition identified in the headspace gas phase of household products, according to product classes. Between 4 (in the product class of nail color removers) and 37 (in the product class of cleaning products) compounds were detected in the headspace gas phase of each class. Several compounds were identified in more than one class. For example, acetone was determined in five of the eight classes (cleaning products, glues, nail color removers, paints, and polishes).

Certain compounds that were present in the cleaning products were identified in cleaning products sold in other countries. For example, five compounds (2-butoxy ethanol, camphene, 3-carene, limonene, β-myrcene, α-pinene, and β-pinene) were identified in certain cleaning products sold in Canada [16] and Korea. However, 2-hexyloxyethanol and α-phellandrene, that were identified in the Canadian products, were not detected in the Korean products. Meanwhile, the number of compounds identified in the cleaning products sold in Korea (37) exceeded the number identified in the cleaning products sold in Canada (8), although the number of cleaning products surveyed in the two studies is not same. Similarly, Zhu et al. [16] identified just one compound, ethyl acetate, in one nail color remover sold in Canada, whereas the current study determined four other compounds (acetone, butyl acetate, cyclohexane, 2,2,4-trimethyl-1,3-dioxolane) in three nail color removers sold in Korea.

3.3. Frequency of occurrence

The frequency of occurrence for each analyte is presented in Table 3. Of the 59 household products analyzed, 58 were found to emit 1 or 72 compounds at chromatographic peak areas above 10^4 and at a spectral search quality of greater than 70%. There were 11 analytes detected at a frequency of more than 10%: limonene (44%), ethanol (31%), acetone (19%), α-pinene (19%), decane (17%), toluene (17%), β-myrcene (12%), *m*,*p*-xylenes (12%), ammonia (10%), ethylbenzene (10%), and

Table 3
Frequency of occurrence for analytes in household products

Chemical	No. products emitting chemical	% of products emitting chemical	Household products emitting chemical
Acetaldehyde	2	3.4	Air freshener, deodorizer
Acetone	11	19	Bleach, laundry detergent, laundry stain remover, floor glue, nail color remover, oil paint, furniture polish
Ammonia	6	10	Disinfectant, dry cleaning, general purpose cleaner, laundry stain remover, oven cleaner, fabric deodorizer
iso-Amyl acetate	1	1.7	Disinfectant
Benzene	2	3.4	Laundry detergent, nail polish
2,3-Butanedione	1	1.7	Wallpaper glue
2-Butanol	1	1.7	Fabric softener
2-Butoxy ethanol	1	1.7	Glass cleaner
Butyl acetate	1	1.7	Nail color remover
Butyl benzene	1	1.7	Oil paint
Butyl cyclohexane	1	1.7	Oil paint
Camphene	3	5.1	Air freshener, disinfectant, oven cleaner
δ-3-Carene	1	1.7	Dry cleaning
Chloroform	4	6.8	Bleach, disinfectant, general purpose cleaners
1-Chloro-2-methylbenzene	1	1.7	Fabric deodorizer
1,8-Cineole	5	8.5	Bleach, disinfectant, general purpose cleaners, glass cleaner, oven cleaner
iso-Cineole	3	5.1	Disinfectant, general purpose cleaners, oven cleaner
Cyclohexane	1	1.7	Nail color remover
Decane	10	17	Air freshener, disinfectant, furniture polish, general purpose cleaners, laundry stain remover, liquid pesticide, oil paint
2,6-Dimethyl-heptane	1	1.7	Oil paint
2,7-Dimethyl-undecane	1	1.7	Furniture polish
1,4-Dioxane	1	1.7	Dishwashing detergent
Dodecane	4	6.8	Air freshener, liquid pesticide, oil paint
Ethanol	18	31	Air freshener, dishwashing detergent, disinfectant, fabric detergent, fabric softener, glass cleaner, laundry detergent, nail polish, oven cleaner
Ethyl acetate	5	8.5	Air freshener, dishwashing detergent, nail polish, wallpaper glue
Ethylbenzene	6	10	Floor glue, oil paint, wallpaper glue, water-based paint
Ethyl cyclohexane	3	5.1	Oil paint
1-Ethyl-3-methyl benzene	1	1.7	Oil paint
1-Hexadecanol	1	1.7	Dishwashing detergent
Hexane	6	10	Air freshener, disinfectant, furniture polish, household sealant
Limonene	26	44	Air freshener, bleach, dishwashing detergent, disinfectant, dry cleaning, fabric deodorizer, fabric softener, furniture polish, general purpose cleaner, glass cleaner, laundry detergent, liquid pesticide, nail polish, oil paint, oven cleaner
cis-Limonene oxide	1	1.7	Bleach
trans-Limonene oxide	1	1.7	Bleach
Methoxy ethane	1	1.7	Fabric softener
2-Methoxy propane	1	1.7	Fabric softener
Methyl acetate	2	3.4	Wallpaper glue
2-Methyl-butane	2	3.4	Liquid pesticide
Methyl cyclobutane	1	1.7	Household sealant
Methyl cyclopentane	1	1.7	Household sealant, oil paint
Methyl ethyl ketone	1	1.7	Household sealant
2-Methyl-nonane	1	1.7	Oil paint
3,7-Methyl-3-octanol	1	1.7	Laundry stain remover
2-Methyl-pentane	1	1.7	Air freshener
3-Methyl-pentane	1	1.7	Disinfectant
4-Methyl-2-pentanone	2	3.4	Oil paint
1-Methyl-propyl benzene	1	1.7	Oil paint

Table 3 (*Continued*)

Chemical	No. products emitting chemical	% of products emitting chemical	Household products emitting chemical
β-Myrcene	7	12	Dishwashing detergent, dry cleaning, fabric deodorizer, furniture polish, general purpose cleaner
Nonadecane	1	1.7	Air freshener
Nonane	1	1.7	Oil paint
Octane	3	5.1	Disinfectant, general purpose cleaner, water-based paint
iso-Octane	1	1.7	Water-based paint
Octyl aldehyde	1	1.7	Dry cleaning
3-Pentanol	1	1.7	Dishwashing detergent
α-Pinene	11	19	Air freshener, bleach, dishwashing detergent, disinfectant, dry cleaning, fabric deodorizer, fabric softener, furniture polish, general purpose cleaner, oven cleaner
β-Pinene	4	6.8	Bleach, disinfectant, fabric deodorizer
1-Propanol	1	1.7	Dishwashing detergent
2-Propanol	1	1.7	Nail polish
Propyl cyclopentane	1	1.7	Oil paint
Sabinene	2	3.4	Dry cleaning, oven cleaner
β-Terpinene	1	1.7	General purpose cleaner
γ-Terpinene	1	1.7	Fabric deodorizer
α-Terpinolene	1	1.7	Oven cleaner
Tetradecane	2	3.4	Dishwashing detergent, liquid pesticide
Toluene	10	17	Disinfectant, floor glue, household sealant, nail polish, oil paint, wallpaper glue
Tridecane	2	3.4	Liquid pesticide
2,2,4-Trimethyl-1,3-dioxolane	1	1.7	Nail color removers
2,2,4-Trimehyl pentane	4	6.8	Oil paint, water-based paint
Undecane	3	5.1	Air freshener, disinfectant, liquid pesticide
o-Xylene	4	6.8	Floor glue, oil paint
m,*p*-Xylene	7	12	Floor glue, oil paint, water-based paint

hexane (10%). In contrast, Sack et al. [1] reported that from the purge-and-trap technique, limonene was not detected in any of 67 households sold in the U.S.A., and that α-pinene was only detected in 5 of 991 households. Three types of terpenes (limonene, α-pinene, and β-myrcene) can potentially react with ozone to generate secondary pollutants with other oxidants such as hydroxyl radicals and nitrogen oxides [12,13]. These terpenes are added to household products due to their favorable odor and solvent properties [2]. Analytes most commonly found in household products sold in the U.S.A. included methylene chloride, 1,1,1-trichloroethane, acetone, 2-butanone, ethylbenzene, methylcyclohexane, *n*-octane, toluene, and xylene [1].

The results of the present study and those of previous studies were compared regarding the presence of 11 analytes which occurred at a frequency of more than 10% in matching household product categories. The household product categories which are reported to emit limonene for those sold in Korea and in other countries include air freshener [15,24], furniture polish [17], disinfectants, general purpose cleaners, and glass cleaners [16]. Furthermore, theses categories include disinfectant and furniture polish for α-pinene [17], glue for xylenes, furniture polish and pesticide for decane [22], furniture polish for hexane [1], and general purpose cleaners for β-myrcene [16]. Meanwhile, it is noteworthy that butoxy ethanol, whose use has raised health concerns [25], exhibited a lower frequency of occurrence (1.7%) in household products and butoxy ethanol was detected in glass cleaner only in the present study. In contrast, butoxy ethanol was detected in several household products, including glass cleaners, sold in Canada [16].

4. Conclusions

The current study evaluated the emissions composition for selected household products currently sold in Korea, using headspace analysis. The chemical composition and concentrations of total VOCs broadly varied across products, even within the same product category. Several compounds were identified in more than one product class. In the current study, the chemical composition of certain household products was different from that of previous studies conducted in other countries.

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