



# Methyl siloxanes in barbershops and residence indoor dust and the implication for human exposures

Nannan Liu <sup>a,b</sup>, Lin Xu <sup>a</sup>, Yaqi Cai <sup>a,c,\*</sup>

<sup>a</sup> State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Science, Chinese Academy of Sciences, Beijing 100085, China

<sup>b</sup> School of Environmental and Municipal Engineering, Tianjin Chengjian University, Tianjin 300384, China

<sup>c</sup> University of Chinese Academy of Sciences, Beijing 100049, China



## HIGHLIGHTS

- The MSs in barbershop dusts were much higher than those from non-occupational sites.
- The MSs in barbershop dusts increased with quantities of daily used PCPs.
- The MSs in household dusts were significantly proportional to the number of occupants.
- The exposure doses of MSs were higher for barbers than those for general adults.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Methyl siloxanes (MSs) have been widely added as emollients or solvents in various personal care products (PCPs) such as soap, shampoo and hair conditioner. A considerable percentage of MSs are released into the atmosphere directly from the use of PCPs in indoor environments. Indoor dust is a major reservoir of MSs, due to their particle-binding affinity. Dust ingestion could be one exposure pathway for special populations (hairdressers, for example). Though the toxicity of MSs is known, there is no information regarding the occurrence of MSs in barbershop dusts and the exposure of hairdressers through indoor dust ingestion. In the present study, the levels of three cyclic siloxanes (D4–D6) and 13 linear siloxanes (L4–L16) were measured in indoor dust samples collected from barbershops, and some other microenvironment including bathhouses, dormitories and households for comparison. The concentrations of  $\sum$  MSs in barbershop dusts were one to two orders of magnitude higher than those from the other three indoor microenvironments. The daily intakes of  $\sum$  MSs through indoor dust ingestion were estimated using the model of worst-case exposure (95th percentile concentration) and high dust ingestion. For the hairdressers in workplaces (barbershops), the exposure rates were 14.3 ng/kg-bw/day, and for the general population, college students and toddlers (1–3 yr), the corresponding exposure rates in living spaces were 3.43, 2.00 and 222 ng/kg-bw/day, respectively. Such high exposure levels of MSs through dust ingestion indicate that we should not overlook the potential health risks for occupational groups and toddlers.

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\* Corresponding author at: State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Science, Chinese Academy of Sciences, Beijing 100085, China.

E-mail address: [caiyaqi@rcees.ac.cn](mailto:caiyaqi@rcees.ac.cn) (Y. Cai).

## 1. Introduction

Methyl siloxanes (MSs) are organo-silicone compounds with –(CH<sub>3</sub>)<sub>2</sub>SiO– structural units. Two major groups of MSs are cyclic and

linear siloxanes ( $C_n$  and  $L_n$  with  $n$  as the numbers of silicon atoms) (Tran and Kannan, 2015). These compounds have been widely used in consumer products and industrial processes for decades due to their excellent properties such as low surface tension, high thermal stability, and lubricating properties (Capela et al., 2016; Homem et al., 2017). For example, MSs are added to personal care products (PCPs) as emollients or solvents in various products throughout the world (Biesterbos et al., 2015; Capela et al., 2016; Montemayor et al., 2012; Wang et al., 2012). MSs were detected in 88% of the 158 PCPs (such as shampoos, hair conditioners, toilet soaps and makeup products) marketed in China, and the highest concentrations of D4, D5, D6 and  $\sum L4-L14$  were 72.9,  $1.11 \times 10^3$ , 367 and  $5.26 \times 10^4$   $\mu\text{g/g}$ , respectively (Lu et al., 2011). In Portugal, MSs ( $0.003-1.20 \times 10^3$   $\mu\text{g/g}$ ) were detected in 96% of 123 personal care products (PCPs), and shampoo exhibited the highest concentration for cyclic siloxanes (Capela et al., 2016). Due to their extensive usage, cyclic volatile methyl siloxanes (cMSs) – D4, D5 and D6 – have been cataloged as high production volume (HPV) chemicals by the Organization for Economic Cooperation and Development and the US Environmental Protection Agency (Hong et al., 2014; Liu et al., 2014). However, with their toxic effects on reproductive, immune, and nervous systems being identified increasingly, the use of MSs has been gradually restricted (McKim et al., 2001; Quinn et al., 2007). For instance, The European Chemicals Agency (ECHA) has made the restriction on D4 and D5 in “wash off” personal care products in 2016 (ECHA, 2016).

In recent years, studies have reported for their occurrence in a wide range of environmental samples, including outdoor air, wastewater, sediment, sewage sludge, and biota, including humans (Homem et al., 2017; Knoerr et al., 2017; Wang et al., 2017; Xu et al., 2015). A considerable percentage of MSs are released into the atmosphere directly from the use of products containing MSs (Biesterbos et al., 2015; Lu et al., 2010). In Europe, the total emission of cVMSs (D4–D6) into the atmosphere was approximately 9 times higher than that into the water through resident's daily activities (Brooke et al., 2009). Moreover, the high particle-binding affinities of MSs made adsorption onto dust and the indoor dust the major reservoir of MSs (Pieri et al., 2013).

In view of their toxic properties, human exposure to MSs has aroused increasing concern (Hori and Kannan, 2008; Lu et al., 2010). Several populations that might be exposed to MSs included occupational workers, consumers, and the general public. Occupational workers consist of persons who work in the production of MSs, in the formulation of MSs into PCPs, and in the use of these products in professional settings, such as beauticians and barbers (Franzen et al., 2016). Dust ingestion was one main human exposure pathway for many volatile organic compounds (VOCs) (Kim et al., 2012; Little et al., 2012). Unfortunately, there are limited studies reporting the population exposure of MSs through dust ingestion, compared with many studies on POPs such as polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) (Mercier et al., 2011; Muenhor et al., 2010; Zhong and Zhu, 2013). Up to now, there are some studies reporting the MSs in indoor dust collected from general population living environment such as residential houses, offices, schools and laboratories. However, the reports on MSs in dust from occupational areas are very limited. Xu et al. (2015) reported that high levels of MSs were detected in dust samples from ordinary locations and industrial plants. Moreover, very little information on the occupational exposure to MSs for those who use PCPs in professional settings was available. Tran and Kannan (2015) detected the highest concentrations of MSs ( $6.21 \mu\text{g/m}^3$ ) in bulk indoor air (particulate plus vapor phases) from 6 hair and nail salons, suggesting that the dust in such microenvironment may contain relatively high concentrations of MSs. Moreover, poor ventilation conditions, high stability of MSs, and long service hours may expose hairdressers to much more MSs than the general public (Franzen et al., 2016). Therefore, determination of MSs in indoor dust from barbershops and evaluation of occupational exposure are imperative to enable risk assessment.

This research focuses on the occurrence and human exposure (occupational exposure for barbers in particular) of MSs in indoor environments. The purposes of the present study were to: (a) determine the levels and compositions of MSs in barbershop dusts and compare them with non-occupational dusts, and (b) estimate the corresponding exposure levels to the general population and hairdressers through indoor dust ingestion. To our knowledge, this is the first study to report the residue levels of MSs in barbershop dust. This study will provide important baseline data for future research on the occurrence and behavior of these compounds in indoor microenvironments.

## 2. Materials and methods

### 2.1. Sampling and preparation

A total of 114 indoor dust samples were collected from barbershops ( $n = 36$ ), university dormitories ( $n = 24$  and 18 for female and male, respectively), urban households ( $n = 30$ , from parlors, bedrooms and bathrooms) and bathhouses ( $n = 6$ ) in Tianjin, China. Sampling was performed through wiping and brushing from no-floor surfaces of furniture. All samples were sieved using stainless steel sieves ( $60 \mu\text{m}$ ) to remove large debris, packed in solvent-washed aluminum foil and sealed in polyethylene zip bags, and then stored at  $-20^\circ\text{C}$  immediately prior to analysis.

Different questionnaires were distributed to the participants. For barbershops, the contents of the questionnaires contained the number of permanent employees, daily business hours, types and amount of hair-personal care products used each day. For households, the questionnaires referred to information about the number of permanent residents in the household and the time since their last clean up.

### 2.2. Reagents, standards and other materials

The analytes in this study included three cyclic siloxanes and 13 linear siloxanes. Cyclic siloxane standards (purity >98%) including D4, D5 and D6, linear siloxane standards (purity >98%) including L4, polydimethylsiloxane mixture (PDMS, L5–L16), and tetrakis (trimethylsilyloxy) silane (M4Q, purity >97%) were purchased from Sigma-Aldrich (St. Louis, MO, USA). Solvents, including methanol, ethyl acetate, n-hexane (HEX) and acetone, were all of HPLC grade and obtained from Fisher Scientific. Anhydrous sodium sulfate ( $\text{Na}_2\text{SO}_4$ ) was baked at  $450^\circ\text{C}$  for 4 h prior to use.

### 2.3. Sample pre-treatment and quantitative analysis

The sonication-assisted extraction procedure referred to in a previous study (Lu et al., 2010) was used with minor modifications. Briefly, dust samples were sieved through a  $500\text{-}\mu\text{m}$  mesh sieve. 0.2 g of the sieved sample was spiked with 100  $\mu\text{L}$  of 1 mg/L M4Q solution (internal standard), vortexed for 5 min at 2500 rpm with 10 mL of ethyl acetate/HEX mixture (V/V, 1:1), and then extracted by sonication for 15 min. The mixture was centrifuged at 3500 rpm for 15 min and the supernatant was collected. The extraction procedure was repeated three times. The total extract was dried by a cartridge filled with 1.0 g of  $\text{Na}_2\text{SO}_4$ , and then the cartridge was eluted with an additional 5 mL of ethyl acetate/HEX mixture (1:1). Finally, the effluent was concentrated to 1 mL under a gentle stream of  $\text{N}_2$  before GC–MS analysis.

### 2.4. Quality assurance and quality control (QA/QC)

Due to the extensive use of MSs in consumer and laboratory products and their broad distribution in environment, we took a series of precautions to avoid contamination during sample collection and analysis: 1) the analyst refrained from using PCPs (cosmetic products and hand cleaners), and powder-free nitrile gloves were used throughout the experiment; 2) before use, all glass tubes were rinsed with HEX, and then baked at  $300^\circ\text{C}$ ; 3) prior to use,  $\text{Na}_2\text{SO}_4$  cartridges were

immersed in hexane for 4 h, and subsequently rinsed with 10 mL of HEX. After rinsing, the cartridges were dried using purified nitrogen; 4) during GC–MS analysis (especially for D4–D6), low injector port temperature (200 °C) and low-bleed capillary columns (HP-5MS) were selected for minimizing targets bleeding from the inlet septum and capillary columns.

Field blanks of dust were prepared with MSs-free Na<sub>2</sub>SO<sub>4</sub>. D4, D5, and D6 detected in field blanks of dust were 0.5–1.0 ng/g, while linear siloxanes were not detected. In the present study, the concentrations of MSs in all samples were corrected with field blanks. Limit of quantifications (LOQs) of cMSs (D4–D6) were determined as 10 times the standard deviations of field blank signals ( $n = 7$ ). For L3–L16 (IMSS for short) not detected in field blanks, the LOQs were determined as 10 times the standard deviation of signals of field blank samples ( $n = 7$ ) spiked with target compounds at known concentrations (3–7 ng/g). LOQs of MSs (D4–D6 and L4–L16) were 0.5–1.2 ng/g for dust, and the average recoveries of MSs for dust were 82% ± 4% to 94% ± 5% (Table S1).

### 2.5. Statistical analysis

For descriptive analysis Microsoft Excel 2007 was used. Statistical analysis was performed using the software OriginPro 9.0 (OriginLab Inc., USA) and SPSS (Version 18.0). Spearman correlation and linear regression analysis were conducted to assess relationships between external factors (such as the quantities of PCPs used daily in indoor environments and the number of occupants, which were obtained from the questionnaires) on the concentrations of MSs in dust samples. The Pearson correlation test was used to assess the correlations between alterations and concentrations of the 16 siloxanes in dust samples. For all comparisons,  $p$ -value of <0.05 was considered statistically significant.

## 3. Results and discussion

### 3.1. Levels of MSs in indoor dust

Sixteen MS compounds [cMSs (D4–D6) and IMSSs (L4–L16)] were detected in 114 samples with the concentrations of  $\sum$  MSs ranging from  $5.29 \times 10^3$  to  $1.33 \times 10^5$  ng/g (mean:  $4.31 \times 10^4$  ng/g) in the barbershop dusts and from 250 to  $8.68 \times 10^3$  ng/g (mean:  $2.50 \times 10^3$  ng/g) in the other three types of non-occupational dusts (Table 1). Relatively higher

**Table 1**  
Summary of measured MSs concentrations (ng/g) in four indoor dust types.

Type (N) <sup>b</sup>		D4	D5	D6	$\sum$ L4–L16 <sup>a</sup>	Total
Barbershops (36)	Mean	3644	8626	1848	29,009	43,126
	Min	51.2	94.0	68.40	3813	5293
	Max	17,055	42,736	8380	81,543	132,599
	Fre%	100	100	100	–	–
Bathhouses (6)	Mean	375	$1.15 \times 10^3$	402	3746	5678
	Min	234	579	225	1039	2131
	Max	584	1534	521	5964	8164
	Fre%	100	100	100	–	–
Dormitory (42)	Mean	55.7	171	87.9	1776	2091
	Min	n.d.	12.5	n.d.	318	334
	Max	251	444	292	7204	7905
	Fre%	95	100	98	–	–
House (30)	Mean	84.0	202	78.9	2084	2449
	Min	n.d.	4.41	3.04	243	250
	Max	383	999	306	7300	8678
	Fre%	93	100	100	–	–
Total (114)	Mean	1212	2901	658	10,560	15,332
	Min	n.d.	4.41	n.d.	243	250
	Max	17,055	42,736	8380	81,543	132,599
	Fre%	96	100	99	–	–

n.d.: not detected.

<sup>a</sup> Full information on L4–L16 concentrations in table S2 in supporting information.

<sup>b</sup> The number of the indoor dust samples.

frequencies of occurrences and concentrations were found for L9–L16 than for cyclic siloxanes (D4–D6) and L4–L8 (Fig. 1 and Table S2). The IMSS proportions of  $\sum$  MSs were 66%–85% (68.9% on average) in the studied dust. Concentrations of total IMSSs (mean:  $1.06 \times 10^4$  ng/g) were 2.2 times higher than those of total cMSs ( $4.78 \times 10^3$  ng/g) in all indoor dust samples. These findings suggested that high-molecular-weight IMSSs are widely present in products used indoors in China, which corresponds well with the previous study on the occurrence of MSs in PCPs from China. Lu et al. detected the levels of MSs (D4–D7 and L4–L14) in 158 PCPs marketed in China and found that IMSSs were prevalent in the most studied products, and that high-molecular-weight IMSSs showed higher concentrations than low-molecular-weight siloxanes in most of the products (Lu et al., 2011). In addition, IMSSs (especially congeners with the number of Si > 6) are less volatile and more prone to adsorbing to dust than cMSs, which might be one of important factors causing the dominance of linear siloxanes in these studied dusts (Homem et al., 2017). Different from the profiles of MSs in indoor dust in this study, higher proportions of cMSs than IMSSs were found in indoor air samples in previous studies. Pieri et al. (2013) found that cMSs (D3–D6, 98% of  $\sum$  MSs) were in obviously lower levels than IMSSs (L2–L5). Similar conclusions were found by the research from New York. cMSs (D3–D7) accounted for 78% of the total MSs (D3–D7 and L3–L11) in the indoor air samples from homes, and 86% of the  $\sum$  MSs in the indoor air samples from 6 salons (Tran and Kannan, 2015).

Among the three cMSs, the mean concentrations in dust samples were in the following decreasing order: D5 ( $2.90 \times 10^3$  ng/g), D4 ( $1.21 \times 10^3$ ) and D6 (658). Overall, D5 was also the predominant siloxane, found at 100% frequency in indoor dust. Indoor dust samples taken in this study also had a pattern similar to those of the personal care products. Personal care products contained D5 at concentrations as high as 14.3% by weight (Horii and Kannan, 2008). Capela et al. (2016) also reported the predominance of D5 in cosmetics and hair care products. In 44 hair care products marked in China, the mean concentrations of MSs were found to be  $13.8 \times 10^3$  ng/g for D4,  $54.2 \times 10^3$  ng/g for D5,  $16.7 \times 10^3$  ng/g for D6 and  $15.7 \times 10^3$  ng/g for  $\sum$  L4–L14, respectively (Lu et al., 2011).

Relationships among the concentrations of 16 MSs in indoor dust collected on four occasions were also examined using the Pearson correlation analysis (Table S3–S5). Most pair-wise comparisons of MSs showed significant correlations (Pearson correlation coefficients ( $r$ ) close to 1,  $p < 0.05$ ), indicating similar sources (such as PCPs) of these compounds in each indoor environment.

### 3.2. Occurrence and profiles of MSs in four different indoor dust

The concentrations of  $\sum$  MSs in dust from barbershops were 1–2 orders of magnitudes higher than those in other three non-occupational indoor dusts (Fig. 2a). The mean levels of  $\sum$  MSs in four different indoor dust followed the order: barbershops ( $4.13 \times 10^4$  ng/g) > bathhouses ( $5.68 \times 10^3$  ng/g) > houses ( $2.45 \times 10^3$  ng/g) > dormitories ( $2.09 \times 10^3$  ng/g). In the present study, the concentrations of MSs in indoor dust from the studied houses and dormitories were slightly higher than the concentrations of  $\sum$  MSs (D4–D7 and L4–L14) in indoor dust from houses (mean:  $1.51 \times 10^3$  ng/g) and dormitory (815 ng/g) in China reported by Lu et al. (2010), while the levels of MSs in indoor dust from the studied barbershops and households in the present study were slightly lower than the concentrations of  $\sum$  MSs (D3–D7 and L3–L11) in the particulate phase indoors from hair and nail salons ( $n = 6$ , mean:  $2.8 \times 10^4$  ng/g) and houses ( $n = 20$ ,  $4.1 \times 10^3$  ng/g) in Albany, USA (Tran and Kannan, 2015). Airborne particles are just one source of indoor dust after deposition, which might lead to somewhat different results on the concentrations of MSs (Xu et al., 2015).

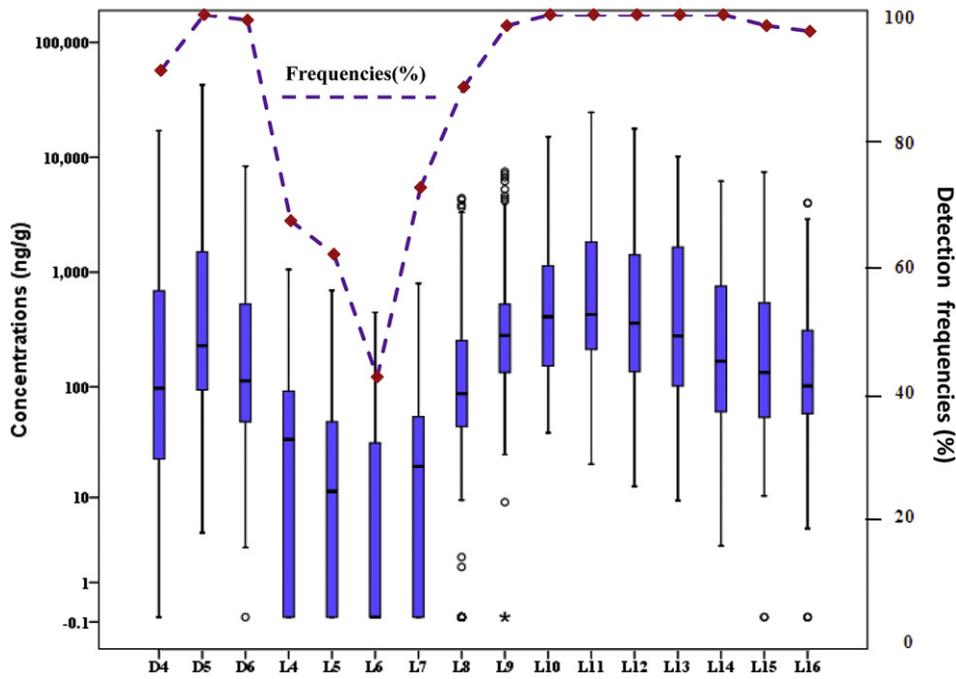


Fig. 1. The detection frequencies and concentrations of methyl siloxanes (MSs) in 114 indoor dusts.

3.2.1. Barbershop dust

The detection frequencies (DFs) of MSs, except for L4–L8, were all 100% in barbershop dust, and the mean concentrations of MSs (except L4–L8 and L16) were all at ppm levels with  $3.64 \times 10^3$ ,  $8.63 \times 10^3$ ,  $1.85 \times 10^3$  and  $2.90 \times 10^4$  ng/g for D4, D5, D6 and  $\sum$  L4–L16, respectively (Table 1). The concentrations of  $\sum$  D4–D6 were the same order of magnitude to those of cMSs ( $\sum$  D4–D6, 2.74–50.4  $\mu$ g/g) in indoor dust from industrial facilities (building, automobile, and textile industries) in previous research (Xu et al., 2015).  $\sum$  IMSs were the dominant targets, accounting for 67% of the  $\sum$  MSs in the barbershop dust averagely. These values were similar to those found in particulate phase of indoor air samples, with  $\sum$  IMSs (L3–L11) accounting for 56% of the  $\sum$  MSs (D3–D7 and L3–L11) (Tran and Kannan, 2015). According to the survey from hairdressers, apart from normal shampoo and hair conditioner, a considerable amount of hair dyes, softeners, perm products, and even makeup products were also consumed in their daily work. A relatively high percentage of occurrence of linear siloxanes was found in skin lotions (89%) makeup/cosmetics products (80%/83%) in previous reports (Horii and Kannan, 2008; Lu et al., 2011). The other products used in barbershops might contain various different blends of MSs from hair

care products, which might lead to the discrepancies between the compositions of MSs in barbershop dusts and in the hair care products reported previously.

Statistically, concentrations of  $\sum$  cMSs and  $\sum$  IMSs significantly increased with the quantities of the consumer products used daily (the sum of all PCPs used daily, according to the questionnaires) in the barbershops (slope = 18,306,  $R^2 = 0.64$  for cMSs and slope = 25,065,  $R^2 = 0.83$  for IMSs,  $p_s < 0.01$ ) (Fig. 3a). Significant and positive correlations were also found between the concentrations of  $\sum$  MSs in dusts and the length of time since the last cleanup (slope = 1133,  $R^2 = 0.38$  for cMSs and slope = 2360,  $R^2 = 0.71$  for IMSs,  $p_s < 0.01$ ) (Fig. 3b). This result indicated that indoor dust could be a potential environmental sink, owing to siloxanes' high particle binding affinities (Kim and Xu, 2016).

3.2.2. Household, bathhouse and dormitory dust

In the dust from 30 households, the mean concentrations of D4, D5, D6 and  $\sum$  IMSs were  $1.21 \times 10^3$ ,  $2.90 \times 10^3$ , 658 and  $1.06 \times 10^4$  ng/g, respectively. The highest  $\sum$  MSs were in bathroom dust, with the mean concentration being  $5.29 \times 10^3$  ng/g, followed by bedroom dust (mean:  $1.61 \times 10^3$  ng/g) and living room dust (mean: 812 ng/g) (Fig. 2b).

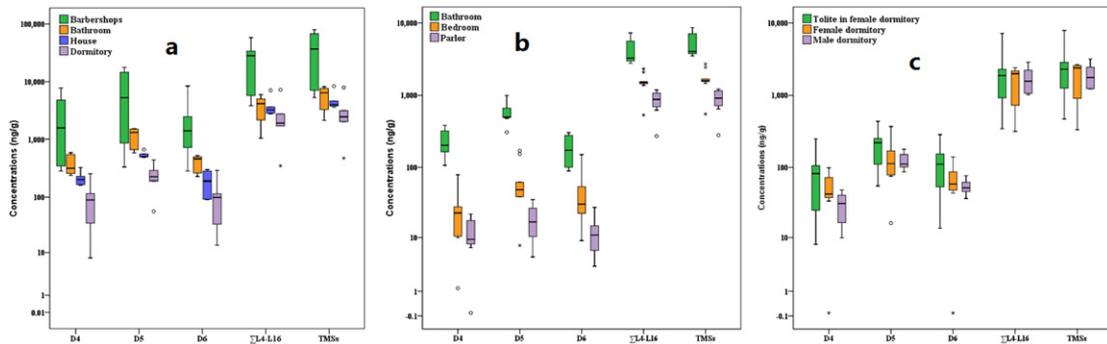
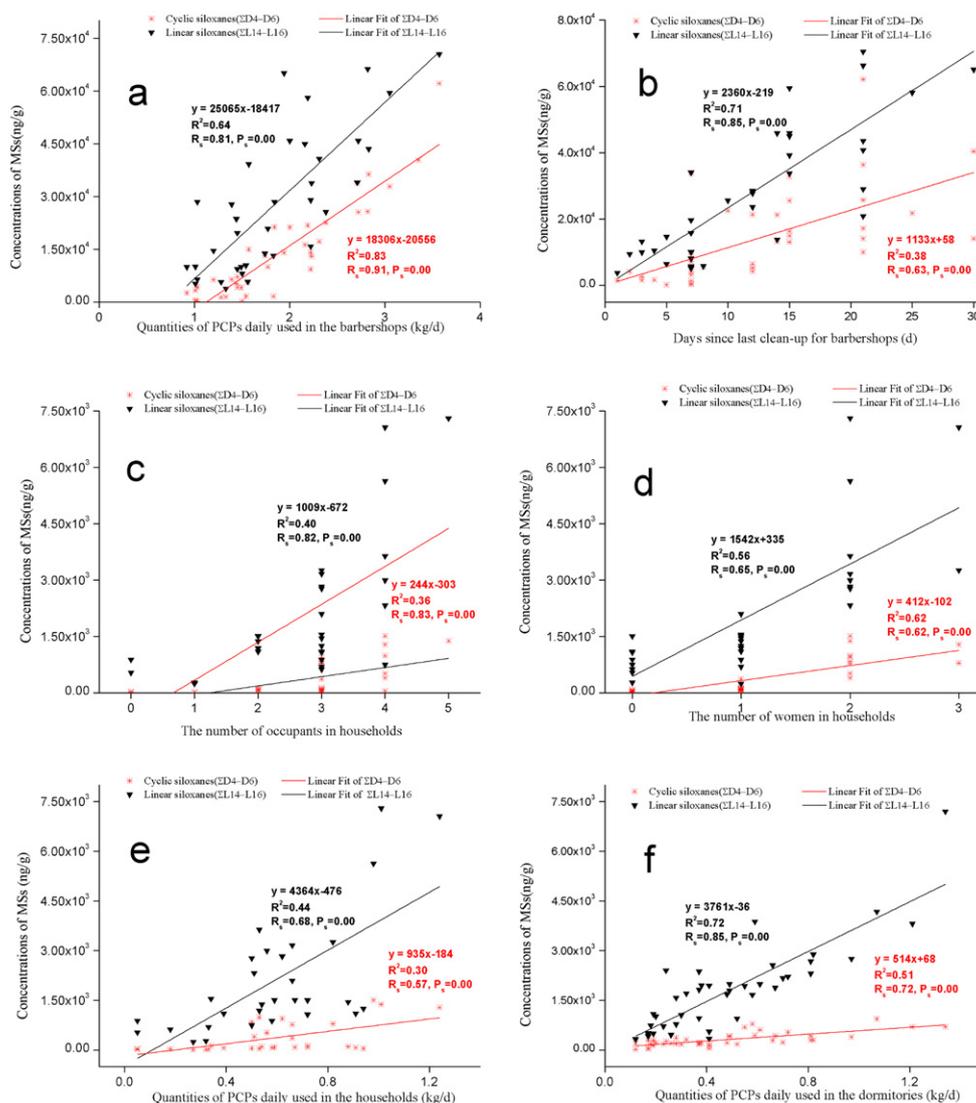


Fig. 2. Box-and-whisker plots of MSs in dust from the indoor microenvironments. a: Barbershops, bathrooms, households and dormitories b: Bathrooms, bedrooms and parlors in households; c: Toilets in female dormitory, female dormitories and male dormitories. The horizontal black line in the box represented the median value and the low and upper edge of the box mark, the 25th and 75th percentiles. The whiskers extending from the box show the lowest and highest values. Values represented by “O” were beyond the 150th percentile of the difference between 25th and 75th percentiles.



**Fig. 3.** Linear regressions and Spearman correlations between mean concentrations of MSs in indoor dusts with some influence factors. a: mean concentrations of MSs indoor dust as related to the quantities of the consumer products daily used in the barbershops. b: mean concentrations of MSs in indoor dust as related to days since last clean up in barbershops. c: mean concentrations of MSs in indoor dust as related to the number of the occupants in household. d: mean concentrations of MSs in indoor dust as related to the number of the women in household. e: mean concentrations of MSs in indoor dust as related to the quantities of the consumer products daily used in households. f: mean concentrations of MSs in indoor dust as related to the quantities of the consumer products daily used in dormitories. R<sup>2</sup> represents correlation coefficient from linear regression; R<sub>s</sub> represents the correlation coefficient for Spearman correlation test; P<sub>s</sub> represents the probability for Spearman correlation to be caused by random sampling.

In this study, the concentrations of MSs in dusts from households were significantly positively related to the number of occupants (slope = 244, R<sup>2</sup> = 0.36 for cVMS and slope = 1109, R<sup>2</sup> = 0.40 for IMSs,  $p_s < 0.01$ ), especially the number of women in the house, which were consistent with preliminary findings (Fig. 3c and d) (Lu et al., 2010). The concentrations of MSs were 912 and 553 ng/g (882 and 535 ng/g for  $\sum$  IMSs in particular) in dust samples from two new houses renovated three months earlier. This result reminded us that the furniture and the decorating materials used in houses might also contain MSs, primarily linear siloxanes, which was consistent with two previous studies from the USA and China. The means of  $\sum$  MSs (D4–D7 and L4–L14) were 93.8 and 15,738  $\mu\text{g/g}$  in sealants (caulking products) and furniture polishes. Another study reported that the concentrations of  $\sum$  D4–D6 ranged from 1.2 to 336  $\mu\text{g/g}$ , and  $\sum$  L5–L16 ranged from 186 to 10,620  $\mu\text{g/g}$  in 24 home paint products sampled from construction sites and paint production plants. (Xu et al., 2015; Horii and Kannan, 2008).

The mean concentrations of total MSs in dormitory dusts collected from seven toilets in female dormitories, 17 female dormitories and 18 male dormitories were  $2.75 \times 10^3$ ,  $2.22 \times 10^3$  and  $1.71 \times 10^3$  ng/g,

respectively (Fig. 2c). The  $\sum$  MSs in dust collected from female bathhouses (mean:  $7.56 \times 10^3$  ng/g) were apparently higher than those from male bathrooms ( $3.80 \times 10^3$ ). The different concentration in indoor dust from female and male dormitories ( $p < 0.05$ ) and bathhouses might be explained by larger usage amount of PCPs among females than males. Consistent with the results of barbershops, the levels of MSs had positive correlations with the PCPs' quantities used daily in the dust samples from households and dormitories (Fig. 3e and f).

### 3.3. Human exposure to MSs via dust ingestion

For adults in general, dermal contact, inhalation and dust ingestion can be considered as potential pathways of exposure to MSs. However, the primary routes of exposure to MSs for hairdressers are still unclear. In this study, the daily intakes (DIs) of MSs by hairdressers in barbershops, and regular adults, college students, and toddlers in living spaces via dust ingestion were calculated. According to previous studies (Lu et al., 2010; Kim et al., 2012), the following typical equation was used.

$$DI_s = C_{dust} \times R_{intake} \times P_{time} / BW \quad (1)$$

**Table 2**

Estimated daily exposure doses (ng/kg-bw/day) of MSs by adults, toddlers (1–3 yr) and hairdressers through dust ingestion.

Environment	Exposure group	Mean-ingestion			High-ingestion		
		5th percentile	Median	95th percentile	5th percentile	Median	95th percentile
House	Adults	0.0479	0.278	1.37	0.120	0.696	3.43
	Toddlers	1.93	11.3	55.5	7.75	45	222
Dormitory	college students	0.0833	0.365	0.799	0.208	0.912	2.00
Barbershop	Hairdressers	0.356	2.67	5.72	0.890	6.68	14.3

where the ' $C_{dust}$ ' is the MS concentrations in indoor dusts (g/g), ' $R_{intake}$ ' means the daily dust intake rate (g/d), and ' $P_{time}$ ' refers to the percentage of the time people spent in different indoor environments (barbershop, home and dormitory). For ' $C_{dust}$ ', three scenarios of MS exposure tested were as following: the low-end exposure (5th percentile MS concentrations), the normal exposure (median MSs concentrations) and the worst-case exposure (95th percentile MS concentrations); ' $R_{intake}$ ' have been reported as 20 and 50 mg/d for adults and toddlers, respectively, while the high intake rates for the two groups were 50 and 200 mg/d (Abdallah and Harrad, 2009; Jones-Otazo et al., 2005). Because of the lack of data on human absorption efficiency about MSs, 100% absorption efficiency was assumed (Lu et al., 2010; Xu et al., 2012); For ' $P_{time}$ ', a typical human time-activity pattern proposed by Klepeis et al. (Klepeis et al., 2001) was adopted in this study, which is 63.8% of the time in home and 22.3% in the office for adults, and 86.1% spent in home for the toddlers. This pattern has already been applied to estimate the human exposure of various organic compounds through dust ingestion/inhalation in various countries (Kim et al., 2012; Lu et al., 2010). ' $BW$ ' was the average body weight (kg). The standard values used were 6 kg for toddlers (1–3 yr) and 72 kg for barbers and college students (U.S. Environmental Protection Agency Child-Specific Exposure Factors Handbook, 2008).

In this study, the high-exposure scenarios of MSs were 14.3 ng/kg-bw/day for hairdressers in workplaces (barbershops), and 3.43, 2.00 and 222 ng/kg-bw/day for regular adults, college students and toddlers (1–3 yr) in living environments (Table 2 and Table S6). The calculated exposure rates of MSs through dust were notably higher than those from one previous report. In Shanghai, the calculated exposure rates of MSs through dust were 21 ng/d for general adults in workplaces and households and 131 ng/d for toddlers in households (Lu et al., 2010). Dust ingestion exposures of MSs calculated in this study (14.3 ng/kg-bw/day for hairdressers) were significantly lower than those through dermal ( $2.5 \times 10^4$  ng/kg-bw/day) and inhalation ( $1.59 \times 10^3$  ng/kg-bw/day) for adults in houses from previous studies, while exposure to MSs through the above three routes was approximately were on an order of magnitude for toddlers (222, 350, and 270 ng/kg-bw/day for dust ingestion, dermal exposure, and inhalation exposure, respectively (Capela et al., 2016; Tran and Kannan, 2015). Dust ingestion could be a relatively more important route of exposure to MSs for toddlers (1–3 yr) than that for adults, since this age group spends much time on the floor, and they are expected not to have extensive use of cosmetics (Lu et al., 2010; Kim et al., 2012). Thus, the available information on the exposure to MSs through house dust should be indispensable to supplement data for other exposure pathways in future research.

#### 4. Conclusion

In this study, the levels of three cyclic siloxanes (D4–D6) and 13 linear siloxanes (L4–L16) were measured in 114 indoor dust samples. These MSs were found to be one or two orders of magnitude higher in barbershop dusts than in other three non-occupational indoor dusts. The congener compositions were similar among all indoor dusts from the four indoor microenvironments: Relatively lower frequencies of occurrences and concentrations were found for L4–L8, than for cyclic siloxanes (D4–D6) and L9–L16 in dust samples. A significant positive correlation existed among MSs in dust samples. Concentrations of MSs

in dust were found to be correlated with the quantities of the consumer products daily used in indoor environments, suggesting that the household PCPs are the major sources of contamination in the indoor micro-environment. The high-exposure scenarios of MSs for hairdressers in barbershops were 14.3 ng/kg-bw/day, and for regular adults, college students and toddlers (1–3 yr), the corresponding exposure rates in living spaces were 3.43, 2.00 and 222 ng/kg-bw/day, respectively, suggesting that the potential health risks existed for occupational groups and toddlers.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2017.09.250>.

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