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# Cyclic and linear siloxanes in indoor air from several northern cities in Vietnam: Levels, spatial distribution and human exposure

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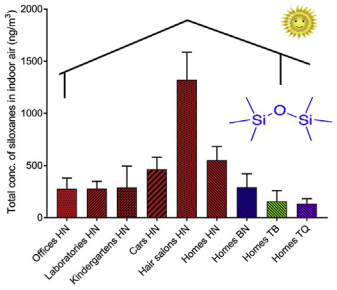
## HIGHLIGHTS

- Siloxanes were found widely in indoor air from Northern in Vietnam.
- Mean concentration of cyclic and linear siloxanes was 228 and 469 ng $m^{-3}$, respectively.
- The highest siloxanes concentrations were measured at hair salons.
- The significant correlation existed between D4, D5, and D6 in indoor air.
- Mean exposure dose to siloxanes through inhalation by infants was 1020 ng kg-$bw^{-1}$ $d^{-1}$.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Earlier studies have reported the occurrence of cyclic and linear siloxanes in personal care and household products. Nevertheless, there is a lack of information on the occurrence of siloxanes in indoor air. In this study, four cyclic and six linear siloxanes were measured in 97 indoor air samples collected from various micro-environments in four cities in northern, Vietnam, during September 2016 to January 2017. The total concentrations of siloxanes (TSi) in particulate and gas phases ranged from 141 to 7220 μg $g^{-1}$ (mean: 1880) and 23.8–1580 ng $m^{-3}$ (mean: 321), respectively. The total concentrations of cyclic siloxanes (TCSi), linear siloxanes (TLSi), and TSi in indoor air were 1.91–1500 ng $m^{-3}$, 21.8–817 ng $m^{-3}$, and 41.8–1950 ng $m^{-3}$, respectively. The highest mean concentration of siloxanes was found in indoor air from hair salons in Hanoi. The concentrations of siloxanes in air collected from homes in Hanoi were higher than those from other smaller cities such as Bacninh, Thaibinh, and Tuyenquang. The human exposures to siloxanes through inhalation were estimated for various age groups based on the measured concentrations. The mean inhalation exposure doses to total siloxanes for infants, toddlers, children, teenagers, and adults were 352, 219, 188, 132, and 95.9 ng kg-$bw^{-1}$ $d^{-1}$, respectively.



## 1. Introduction

Siloxanes are organosilicon compounds containing carbon-

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silicon bonds. Siloxanes are widely used in many products due to their low surface tension, high thermal stability, and smooth texture. Personal care and household products contain siloxanes typical in the order of several percentages by weight (Horii and Kannan, 2008; Wang et al., 2009). Occurrence of octamethylcyclotetrasiloxane (D4; 72.9 μg $g^{-1}$) decamethylcyclopentasiloxane (D5, 1110 μg $g^{-1}$) and total linear siloxanes (L4-L14; 1.02 mg $g^{-1}$) was reported in shampoos and hair conditioners (Lu et al., 2011). Siloxanes also were reported to occur in silicon rubber products (Kawamura et al., 2001), electrical devices, healthcare products, cosmetics, cookware, sealants, and household cleaning products (Watts et al., 1995; Environment Canada, 2011).

The environmental occurrence and distribution of siloxanes were reported in several previous studies (Xu et al., 2015). The mean concentration of total siloxanes (5 cyclic and 15 linear) in sludge samples collected from wastewater treatment plants in South Korea was 45.7 μg $g^{-1}$ (Lee et al., 2014). Cyclic and linear siloxanes were found in sediment and wastewater collected from China (Zhang et al., 2011), Canada (Wang et al., 2013), and Spain (Sanchís et al., 2013). Owing to their widespread use in consumer products, there is a great potential for the occurrence of elevated concentrations of siloxanes in indoor environments (Brooke et al., 2009a, b, c). Few studies have reported the occurrence of siloxanes in the indoor environment. A median total concentration of 5 cyclic and 11 linear siloxanes were determined in indoor dust samples from twelve countries (Tran et al., 2015). Similarly, mean concentration of total siloxanes (5 cyclic and 9 linear siloxanes) in indoor air from Albany, NY, USA, was reported to range from 249 to 6210 ng $m^{-3}$ (Tran and Kannan, 2015). Furthermore, indoor air samples collected from Chicago contained a median concentration of the sum of D4, D5, and dodecamethylcyclohexasiloxane (D6) at 2200 ng $m^{-3}$ (Yucuis et al., 2013).

Siloxanes have been reported to be reproductive and endocrine toxicants (McKim et al., 2001; He et al., 2003; Meeks et al., 2007; Quinn et al., 2007a, b; Siddiqui et al., 2007; Reddy et al., 2007, 2008). Dekant and Klaunig (2016) reported that exposure to D5 decreased release of prolactin. The liver weight increases observed in several toxicity studies with D5 of shorter duration and in the two-year chronic bioassay, D5 inhalation caused a small, but (borderline) statistically significant increase in the incidence of uterine adenocarcinoma at the highest exposure concentration of 160 ppm (Dekant and Klaunig, 2016). A risk assessment conducted in Canada indicated that D5 met the criteria for environmental persistence (Environment Canada, 2011). The environmental exposures and risks associated with siloxanes have been under scrutiny by several environmental and public health agencies in various countries (Surita and Tansel, 2014; Tran et al., 2015). Despite this, there is a lack of information with regard to the sources of human exposure to siloxanes. In this study, we surveyed the occurrence and distribution of four cyclic and six linear siloxanes in 97 indoor air samples collected from four northern cities in Vietnam: Hanoi, Bacninh, Thaibinh, and Tuyenquang. The exposure doses to siloxanes through inhalation were estimated for infants, toddlers, children, teenagers, and adults based on the measured siloxanes concentrations in indoor air.

## 2. Materials and methods

### 2.1. Standards and solvents

Hexamethylcyclotrisiloxane (D3), octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), undodecamethylcyclohexasiloxane (D6), decamethyltetrasiloxane (L4) and dodecamethylpentasiloxane (L5) with a purity of >97%, were purchased from Sigma-Aldrich (St.Louis, MO, USA). PDMS 200 fluid (viscosity of 5 cSt) that contained other linear polydimethyl siloxanes (L6, L7, L8, and L9) was purchased from Sigma-Aldrich. Compositions of siloxanes in PDMS 200 fluid were determined by a gas chromatography with a flame ionization detector. The compositions of L6, L7, L8, and L9 were respectively 17.5; 18.4; 16.5; and 14.3%. Tetrakis (trimethylsiloxy)-silane (M4Q) of 97% purity was from Aldrich, and this compound was used as a surrogate standard for all target compounds. All standards were dissolved in hexane. Hexane and acetone (for analysis) were purchased from Merck KGaA (Darmstadt, Germany), and dichloromethane (analytical reagent grade) was purchased from Fisher Scientific (Leicestershire, UK).

### 2.2. Sample collection

Polyurethane foam (PUF) plugs (ORBO-1000, PUF dimension: 2.2 cm O. D x 7.6 cm length) were purchased from Supelco (Bellefonte, PA, USA). For the analysis background levels of siloxanes, two PUF plugs were extracted twice with 100 mL mixture of dichloromethane (DCM) and *n*-hexane (3:2, v:v) and analyzed by gas chromatography-mass spectrometry (GC-MS). Each of the newly purchased PUF plugs contained D4, D5, and D6 at 5.0–12.7, 3.1–27.5, and 3.5–22.9 ng, respectively (n = 7). Therefore, all PUF plugs required additional purification prior to use. PUFs were purified by shaking with 100 mL mixture of DCM and *n*-hexane (3:2, v:v) for 20 min. The procedure was performed twice. The cleaned PUFs were wrapped in aluminum foil, stored in a glass jar, and kept in an oven at 100 °C until sampling. The quartz fiber filters (Whatman, grade MQ-A, pore size: 2.2 μm with a particle retention rating at 98% efficiency in liquid, 32 mm diameter) were prepared by heating at 400 °C for 24 h. The purified quartz fiber filters were kept in an oven at 100 °C until use. The quartz fiber filters were weighed in an analytical balance (ES 225MS-DR, Switzerland, to nearest 0.01 mg) before and after the collection of air samples for the determination of particle content. Two PUF plugs were packed in tandem in a glass tube (ACE glass, 2,2 cm O. D x 25 cm length), and the quartz fiber filter was held with a Teflon cartridge (Supelco, PUF filter cartridge assembly) on top of the glass tube packed with PUF plugs.

Indoor air samples were collected for 12–24 h by a low-volume air sampler (LP-7 230 V pump kit, A.P. Buck Inc., Orlando, FL, USA) at a flow rate of 4 L per min. The total volume of air collected from each location ranged from 2.88 to 5.76 $m^3$. After collection, samples were immediately wrapped with aluminum foil and transferred to the laboratory. Air samples (both PUFs and filters) were kept for no longer than 2 weeks at −18 °C until analysis. Indoor samples were collected during September 2016 to January 2017 (Table S1, Supporting Information). The sampling locations in Hanoi city were grouped into six categories as homes (n = 19), cars (n = 8), kindergartens (n = 7), laboratories (n = 19), offices (n = 9), and hair salons (n = 13). For other cities, air samples were only collected at homes: Bacninh (n = 8), Thaibinh (n = 6), and Tuyenquang (n = 8). Most types of room for sampling at homes are living rooms. (Table S2 and Fig. S1, Supporting Information). Hanoi is the metropolitan center in Vietnam with the most population density (2170 people $km^{-2}$), followed by Bacninh (1400 people $km^{-2}$), Thaibinh (1140 people $km^{-2}$), and Tuyenquang (130 people $km^{-2}$) (Vietnam Government, General Statistic Office, 2015) (Table S2, Supporting Information).

### 2.3. Sample preparation

Two hundred nanograms of M4Q was spiked as a surrogate standard into every sample (both PUFs and filters). The extraction procedure was similar to that described earlier (Tran and Kannan, 2015) with minor modifications. Two PUF plugs were extracted