Chemosphere 212 (2018) 330–336

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Cyclic siloxanes in indoor environments from hair salons in Hanoi, Vietnam: Emission sources, spatial distribution, and implications for human exposure

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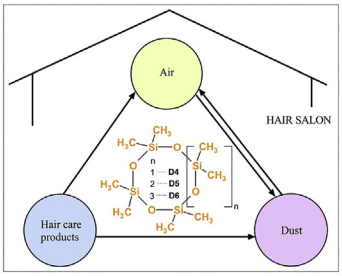
HIGHLIGHTS

- Total concentration of cyclic siloxanes in hair care products were up to 515 μg g^{-1}.
- High levels of CSis were found in indoor air samples.
- D5 had strong concentration correlation between indoor air and dust samples collected from hair salons in Hanoi, Vietnam.
- Estimated exposure doses to total CSis through inhalation for woman and man were respectively 103 and 79.5 ng kg-bw^{-1} d^{-1}.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 18 May 2018
Received in revised form
16 August 2018
Accepted 19 August 2018
Available online 20 August 2018

Handling Editor: Andreas Sjodin

Keywords:
Cyclic siloxanes
Hair salons
Hair care products
Indoor environments
Correlation

ABSTRACT

In this study, three typical cyclic siloxanes (CSis) were found in hair care products, indoor dust and indoor air samples at hair salons in Hanoi, Vietnam. The total concentrations of CSis in three kinds of hair care products ranged from 8.77 to 515 μg g^{-1}. The mean and median concentrations of CSis in dust samples collected at hair salons were 671 and 654 μg g^{-1}, respectively. The total concentrations of CSis in indoor air samples collected at the hair salons ranged from 415 to 2610 ng m^{-3} (mean: 1030; median: 849 ng m^{-3}, respectively). Among three cyclic siloxanes mentioned in this study, decamethylcyclopentasiloxane (D5) was found at the highest level in all categories of samples. D5 had also a strong correlation between indoor dust and indoor air from hair salons (the coefficient of determination (R^2): 0.852); meanwhile dodecamethylcyclohexasiloxane (D6) and octamethylcyclotetrasiloxane (D4) had good correlations ($R^2 = 0.618$ and 0.585, respectively). This result indicates that hair care products are main emission source of cyclic siloxanes in indoor environments at the hair salons. The average exposure doses to total SCis through both of dust ingestion and inhalation were estimated to be 103 and 79.5 ng kg-bw^{-1} day^{-1} for women and men, respectively. These levels were higher than those reported for some Asian countries. This is among the most comprehensive investigations on the emission sources and distributions of cyclic siloxanes in indoor dust and indoor air at hair salons in Vietnam.

1. Introduction

Siloxanes including cyclic siloxanes possess many useful

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https://doi.org/10.1016/j.chemosphere.2018.08.101
0045-6535/

properties such as smooth texture, low surface tension, and high thermal stability, therefore, they are commonly used in the personal care products (PCPs). Some earlier surveys reported the significant amount of cyclic siloxanes contained in PCPs (Loretz et al., 2006, 2008; Horii and Kannan, 2008; Wang et al., 2009). Lu et al. (2011) reported that the concentrations of octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), and dodecamethylcyclohexasiloxane (D6) in hair care products which collected from China were found up to 1110 μg g^{-1} (Lu et al., 2011).

Owing to widespread use of siloxanes in the consumer products, their occurrence and distribution have reported in a wide range of environmental samples such as indoor dust, indoor air, water, and sewage sludge (Lu et al., 2010; Zhang et al., 2011; Sanchís et al., 2013; Wang et al., 2013; Lee et al., 2014, 2018; Tran et al., 2016). Several previous studies have showed that the high levels of siloxanes were found in the indoor air samples collected in the United States of America (Yucuis et al., 2013; Tran and Kannan, 2015), and Italy and the United Kingdom (Pieri et al., 2013). Tran et al. (2015) was mentioned in the previous study, cyclic and linear siloxanes were distributed widely in indoor dust which were collected from twelve countries and the total concentrations of siloxanes measured in indoor dust samples from Kuwait and Greece were as high as 42,800 ng g^{-1} and 25,000 ng g^{-1}, respectively (Tran et al., 2015). In further research, cyclic (D4-D6) and linear (L5-L16) siloxanes were found in plasma samples of industrial workers in China up to 252 ng mL^{-1} (Xu et al., 2015).

In recent years, cyclic siloxanes have actually been interested because of their reproductive and endocrine effects found for animals in laboratory (McKim et al., 2001; Burn-Naas et al., 2002; Siddiqui et al., 2007; Quinn et al., 2007a; Reddy et al., 2008). The uterine wet weights and uterine peroxidase activity were significantly increased levels of D4 exposed in mice (He et al., 2003). Meeks et al. reported that a single 6 h exposure of D4 on the day prior to mating resulted in a significant reduction in fertility of the female rat (Meeks et al., 2007). Previous studies had pointed out that D4 activated the reporter gene at 10 microM, while D5 was considered negative in the estrogen reporter gene assay (Quinn et al., 2007a, b). Recently, Dekant and Klaunig (2016) also reported that the liver effects induced by D5 were consistent as a weak “phenobarbital-like” inducer of xenobiotic metabolizing enzymes and these effects are considered to be an adaptive response (Dekant and Klaunig, 2016). An environmental risk assessment study conducted in the UK suggested that D4 has a long-term fish no-observed-effect concentration (NOEC) of 4.4 μg L^{-1}. A fish NOEC with 4.4 μg L^{-1} in the 14-day prolonged acute toxicity study and a long-term NOEC of 7.9 μg L^{-1} from the Daphnia magna reproduction study (Brooke et al., 2009c). Generally, it was showed that D4 meets the toxic criterion; meanwhile D5 and D6 do not meet the screening for the toxicity (Brooke et al., 2009a, b, c).

The occurrence and distribution of siloxanes in various micro-environments in several cities in northern areas in Vietnam have been recently investigated (Tran et al., 2015, 2016). Our results indicated higher levels of cyclic and linear siloxanes in indoor air from hair salons compared to other micro-environments (Tran et al., 2017). This suggests that hair salons could be a potential environment for human exposure to siloxanes. In order to further clarify the relationship between the use of siloxanes in commercial hair care products and their distribution in environments, in this survey we determined the levels of cyclic siloxanes in hair care products, indoor dust, and indoor air samples collected at hair salons. The concentration correlations of cyclic siloxanes between indoor dust and indoor air samples, and the human exposure to these chemicals for adults worked in hair salons were also considered.

2. Materials and methods

2.1. Solvents and standards

Acetone and *n*-hexane (for analysis) were purchased from Merck KGaA (Darmstadt, Germany), and dichloromethane with a purity of above 99.8% was purchased from Deajung chemicals & metals co., ltd. (Gyonggi-do, Korea). Three cyclic siloxanes including octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), dodecamethylcyclohexasiloxane (D6), and tetrakis(-trimethylsiloxy)silane (M4Q) with a purity of >97%, were purchased from Sigma-Aldrich (St.Louis, MO, USA). The M4Q was used as a surrogate compound for three cyclic siloxanes. All standard and surrogate compounds were dissolved in *n*-hexane with different concentrations from 1 ng mL^{-1} to 1000 ng mL^{-1}.

2.2. Collection and preparation of samples

2.2.1. Hair care products samples

Shampoo and hair gel samples were purchased from retail stores in Hanoi, Vietnam as well as collected directly from hair salons from June to December 2017. Samples were kept at room temperature until analysis (before expired date).

Three hundred nanogram of M4Q was spiked as a surrogate compound into 200–500 mg of hair care product samples in glass tubes. The extraction procedure was performed similarly to earlier studies (Horii and Kannan, 2008; Lu et al., 2011) with minor modification. Samples were shaken with 5 mL of ethyl acetate and *n*-hexane (2:3, v:v) for 10 min and centrifuged (Laboratory Centrifuge LC-04P-L, Zenith Lab Inc.) at 2500 rpm for 5 min. The solvent layer was transferred to another glass tube. The extraction was repeated twice with 3 mL of ethyl acetate and *n*-hexane for each extraction. The total extracted solution was concentrated to about 3 mL by a gentle stream of nitrogen and then passed through anhydrous sodium sulfate (2.5 g) and a PTFE membrane filter (Whatman Uniflo, pore size: 0.22 μm). The final volume was concentrated exactly to 1 mL and transferred into a GC vial, prior to GC-MS analysis.

2.2.2. Indoor dust samples

Indoor dust samples were collected by using a direct broom on the floor and surface of the furniture at hair salons in Hanoi, Vietnam from June to December 2017. Dust samples were packed with aluminum foil and transferred to laboratory. In the laboratory, dust samples were homogenized and sieved through a sieve of 150 μm. Samples were stored in a dark glass jar and sealed at 4 °C until analysis.

300 ng of M4Q was spiked into 400–500 mg of dust sample. The extraction procedure was similar to our previous study with slight modification (Tran et al., 2015). Target compounds were extracted with 5 mL of dichloromethane (DCM) and *n*-hexane (3:2; v:v) by shaking in an orbital shaker (Stuart, Japan) for 5 min. Extraction was repeated more twice. The extracts were evaporated to 1 mL under a gentle stream of nitrogen and then filtered through a PTFE membrane filter (Whatman Uniflo, pore size: 0.22 μm), and transferred into a GC vial.

2.2.3. Indoor air samples

At the same places for dust sampling (Table S1, Supplemental Information), 24 indoor air samples were collected from 20 to 24 h by a low pump (LP-7; AP. Buck Inc., Orlando, FL, USA). The flow rate was set at 5 liter min^{-1} and the air volume was collected in a range from 5.0 to 7.2 m^3. Indoor air samples were collected on two phases (vapor and particulate phases). The sampling method was implemented following some earlier studies with little modification