Science of the Total Environment 537 (2015) 304–308

Contents lists available at ScienceDirect

Science of the Total Environment

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

Occurrence of benzophenone-3 in indoor air from Albany, New York, USA, and its implications for inhalation exposure

Yanjian Wan [a,b], Jingchuan Xue [a], Kurunthachalam Kannan [a,c,*]

[a] *Wadsworth Center, New York State Department of Health, and Department of Environmental Health Sciences, School of Public Health, State University of New York at Albany, Empire State Plaza, P.O. Box 509, Albany, NY 12201-0509, United States*

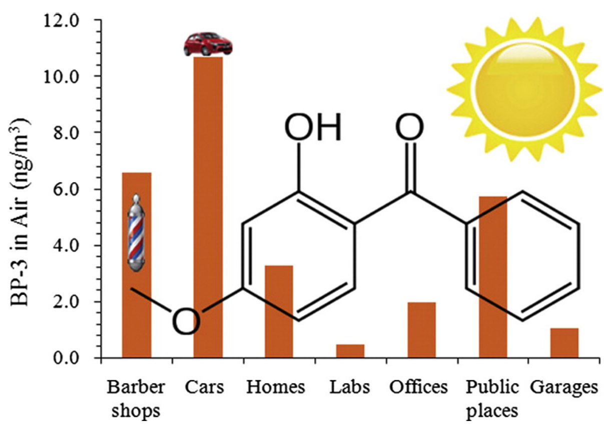
[b] *Center for Disease Control and Prevention of Yangtze River Administration and Navigational Affairs, General Hospital of the Yangtze River Shipping, Wuhan 430019, China*

[c] *Biochemistry Department, Faculty of Science and Experimental Biochemistry Unit, King Fahd Medical Research Center, King Abdulaziz University, Jeddah 21589, Saudi Arabia*

HIGHLIGHTS

- Benzophenones were determined in 81 indoor air samples.
- Benzophenone-3 ranged from 0.19 to 72.0 ng/m^3 with the highest levels in cars.
- Inhalation exposure dose to benzophenone-3 ranged from 0.42 to 1.50 ng/kg-bw/d.
- The contribution of inhalation to total benzophenone-3 intake was <5%.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 25 June 2015
Received in revised form 30 July 2015
Accepted 5 August 2015
Available online xxxx

Editor: Adrian Covaci

Keywords:
Benzophenone-3
Indoor air
Human exposure
UV filter
Cosmetics

ABSTRACT

Benzophenone-3 (BP-3) is a widespread environmental contaminant and an estrogenic compound. Very little is known with regard to the occurrence in indoor air and the inhalation exposure of humans to BP-3. In this study, 81 indoor air samples were collected from various locations in Albany, New York, USA, in 2014 and analyzed for BP-3 by high performance liquid chromatography-tandem mass spectrometry (HPLC-MS/MS). BP-3 was found in all indoor air samples and the overall concentrations in bulk air (vapor plus particulate phases) were in the range of 0.19–72.0 ng/m^3 (geometric mean: 2.67 ng/m^3). The highest concentrations (geometric mean: 10.7 ng/m^3) were found in cars, followed by barber shops (6.57) > public places (5.75) > homes (3.27) > offices (1.96) > garages (1.04) > laboratories (0.47). The estimated geometric mean daily intake (EDI) of BP-3 for infants, toddlers, children, teenagers, and adults through indoor air inhalation from homes was 1.83, 1.74, 1.18, 0.69, and 0.51 ng/kg-bw/day, respectively. Although high concentrations of BP-3 were measured in some microenvironments, the estimated contribution of indoor air to total BP-3 intake was <5% of the total BP-3 intake in humans. This is the first survey on the occurrence of BP-3 in indoor air.

* Corresponding author at: Wadsworth Center, Empire State Plaza, P.O. Box 509, Albany, NY 12201-0509, United States.
E-mail address: kurunthachalam.kannan@health.ny.gov (K. Kannan).

http://dx.doi.org/10.1016/j.scitotenv.2015.08.020

1. Introduction

Benzophenone-3 (2-hydroxy-4-methoxybenzophenone; BP-3) occurs naturally in some plants (IARC, 2012). BP-3 and its derivatives (e.g., BP-1, BP-8) are also commercially manufactured for use as a sunscreen in skin lotions, perfumes, and cosmetics to prevent UV-light from damaging scents and colors in such products (Liao and Kannan, 2014; Kim and Choi, 2014). BP derivatives are also used as UV-light stabilizers in plastic surface coatings and in polymers (Suzuki et al., 2005). Additionally, BP derivatives are used as UV-curing agents in sunglasses, and laundry and household cleaning products (IARC, 2012). Due to the extensive use, BP-3 is a widespread environmental contaminant and has been detected in the environment (Kim and Choi, 2014) and biological samples including human urine (Xue et al., 2015; Gao et al., 2015), serum (Hines et al., 2015; Tarazona et al., 2013), breast milk (Rodriguez-Gomez et al., 2015; Ye et al., 2008), amniotic fluid (Philippat et al., 2013), adipose fat (Wang et al., 2015) and placental tissue (Vela-Soria et al., 2011). Occurrence of BP-3 in 98% of urine samples collected from the United States population suggested that human exposure to this compound is widespread (Calafat et al., 2008).

BP-3 has been reported to elicit estrogenic and anti-androgenic activities (Schlumpf et al., 2001; Schreurs et al., 2005; Suzuki et al., 2005) in laboratory studies. Hormonal balance and reproductive performances were affected by exposure to BP-3 in laboratory animals (Ozaez et al., 2014). Exposure to BP-3 derivatives in women has been associated with higher odds of developing endometriosis (Kunisue et al., 2012). A recent epidemiological study reported that elevated exposure of men to BP-type UV filters diminished couples' fecundity, resulting in a longer time to achieve pregnancy in women (Buck-Louis et al., 2014).

Assessing the sources of human exposure to BP-3 is a subject of considerable interest, if we have to devise solutions to mitigate exposures. Thus far, BP-3 has been reported to occur in personal care products (Liao and Kannan, 2014), sediments and sewage sludge (Zhang et al., 2011), surface water (Poiger et al., 2004; Tsui et al., 2014), drinking water (Diaz-Cruz et al., 2012), foodstuffs (Balmer et al., 2005; Fent et al., 2010), and indoor dust (Wang et al., 2013). However, little is known on human exposure to BP-3 through inhalation of indoor air. Indoor air is a significant source of human exposure to contaminants such as polybrominated diphenyl ethers, perfluoroalkyl sulfonamides, siloxanes, and phthalates (Shoeib et al., 2004; Tran and Kannan, 2015a; Tran and Kannan, 2015b; Ma et al., 2014). Because of the widespread use of BP-3 in household as well as personal care products, this compound is expected to occur in indoor air, and inhalation can be an important route of human exposure. In this study, we conducted a survey of BP-3 in 81 indoor air samples collected in Albany, New York, USA (7 cars, 5 hair salons, 13 homes [day and night], 13 offices, 12 laboratories, 13 public places and 5 automobile [4 repairing, 1 parking] garages). BP-3 exposures via indoor air inhalation for various age groups (infants, toddlers, children, teenagers, and adults) were calculated on the basis of the measured concentrations. This is the first study to report the occurrence of BP-3 in indoor air.

2. Materials and methods

2.1. Chemicals

BP-3 (98%), 2,4-dihydroxybenzophenone (BP-1 or UV-0, CAS# 131-56-6, 99%), 2,2′-dihydroxy-4-methoxybenzophenone (BP-8 or UV-24, CAS# 131-53-3, 98%), 2,2′,4,4′-tetrahydroxybenzophenone (BP-2, CAS# 131-55-5, 97%), and 4-hydroxybenzophenone (CAS# 1137-42-4, 98%) were purchased from Sigma-Aldrich (St. Louis, MO, USA). Isotopically-labeled BP-3 ($^{13}C_6$-BP-3, 99%, CLM-8525) was purchased from Cambridge Isotope Laboratories (Andover, MA, USA). Methanol and ethyl acetate were purchased from J. T. Baker (Phillipsburg, NJ, USA).

2.2. Sample collection and preparation

Pre-cleaned polyurethane foam (PUF) plugs (ORBO-1000 PUF dimensions: 2.2 cm O.D × 7.6 cm length) were purchased from Supelco (Bellefonte, PA, USA). To test the background levels of BP-3, new PUF plugs (purchased from the vendor) were extracted with ethyl acetate and analyzed by liquid chromatography-tandem mass spectrometry (LC-MS/MS). It was found that each pair of newly purchased PUF plugs contained 3.59 ± 1.68 ng BP-3 ($n = 5$). Therefore, all PUF plugs required additional clean-up prior to use. PUF plugs were cleaned up by shaking with 100 mL of ethyl acetate for 30 min, twice. The cleaned PUFs were stored in a glass bottle, and kept in an oven at 100 °C until use. The quartz fiber filters (Whatman, grade QM-A, pore size: 2.2 μm with a particle retention rating at 98% efficiency in liquid, 32 mm diameter) were cleaned, and weighed before and after the collection of air samples as described earlier (Tran and Kannan, 2015b).

Two PUF plugs and the quartz fiber filter were assembled in a glass tube as described earlier (Tran and Kannan, 2015b). All glassware used in sampling and analysis was rinsed with ethyl acetate and methanol and kept at 450 °C immediately prior to use. Indoor air samples were collected for 3 to 24 h by a low-volume air sampler (LP-20; A.P. Buck Inc., Orlando, FL, USA) at a flow rate of 5 L/min. Air samples (both PUFs and quartz fiber filters) were kept in glass bottles for no longer than 2 days prior to analysis. The samples were collected from September to December 2014 at several locations in Albany, New York, USA. The sampling locations were grouped into 7 categories: homes, offices, laboratories, cars, barber shops, automobile garages, and public places (e.g., shopping malls). Prior to analysis, samples (both PUFs and filters) were spiked with 10 ng of $^{13}C_6$-BP-3 as an internal standard. PUF plugs were extracted by shaking in an orbital shaker (Eberbach Corp., Ann Arbor, MI, USA) with 100 mL ethyl acetate for 30 min, twice. The particulate samples were extracted with ethyl acetate by shaking for three times, 5 min every time. The extracts were concentrated in a rotary evaporator at 40 °C to approximately 5 mL, transferred to a 12-mL glass tube and concentrated by a gentle stream of nitrogen to exactly 1 mL, and then transferred into a glass vial.

Table 1
Concentrations of BP-3 in the particulate phase (μg/g), vapor phase (ng/m^3) and bulk air (ng/m^3) of indoor air samples collected from various locations in Albany, New York, USA in 2014.

Locations	n	Particulate phase (μg/g)			Vapor phase (ng/m^3)			Bulk air (ng/m^3)		
		*GM	Median	Range	GM	Median	Range	GM	Median	Range
Barber shops	5	29.4	14.6	8.64–189	3.96	3.42	1.90–13.7	6.57	5.39	2.95–19.4
Cars	7	66.4	43.9	17.9–477	5.28	13.3	0.20–70.0	10.7	18.3	1.57–72.0
Homes	26	32.5	31.2	4.69–205	1.53	1.64	0.07–18.0	3.27	2.92	1.15–24.7
Labs	12	21.4	21.7	5.69–218	0.05	0.08	<LOQ–0.20	0.47	0.42	0.19–4.06
Offices	13	61.9	70.7	13.1–171	0.51	0.59	0.04–9.18	1.96	2.09	0.43–11.9
Public places	13	50.7	48.8	21.5–147	1.50	1.18	0.39–12.2	5.75	4.71	2.57–13.9
Garages	5	8.81	8.15	3.58–23.2	0.56	0.58	0.30–0.88	1.04	0.85	0.59–2.74
Total	81	35.5	32.0	3.58–477	0.85	0.93	<LOQ–70.0	2.67	2.91	0.19–72.0

* GM = geometric mean.