Environment International 112 (2018) 85–99

Contents lists available at ScienceDirect

Environment International

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

Distribution, variability, and predictors of urinary bisphenol A levels in 50 North Carolina adults over a six-week monitoring period

Marsha K. Morgan[a,*], Maliha Nash[b], Dana Boyd Barr[c], James M. Starr[a], M. Scott Clifton[a], Jon R. Sobus[a]

[a] United States Environmental Protection Agency, National Exposure Research Laboratory, Research Triangle Park, NC 27711, USA
[b] United States Environmental Protection Agency, National Exposure Research Laboratory, Las Vegas, NV 89154, USA
[c] Department of Environmental Health, Rollins School of Public Health, Emory University, Atlanta, GA 30322, USA

ARTICLE INFO

Keywords:
Bisphenol A (BPA)
Adults
Urine
Homes
Biomonitoring
Temporal
Determinants

ABSTRACT

Bisphenol A (BPA) is commonly manufactured to make polycarbonate plastics and epoxy resins for use in consumer products and packaged goods. BPA has been found in several different types of environmental media (e.g., food, dust, and air). Many cross-sectional studies have frequently detected BPA concentrations in adult urine samples. However, limited data are available on the temporal variability and important predictors of urinary BPA concentrations in adults. In this work, the major objectives were to: 1) quantify BPA levels in duplicate-diet solid food, drinking water, hard floor surface wipe, and urine samples (first-morning void [FMV], bedtime, and 24-h) collected from adults over a six-week monitoring period; 2) determine the temporal variability of urinary BPA levels using concentration, specific gravity (SG) adjusted, creatinine (CR) adjusted, and excretion rate values, and; 3) examine associations between available study factors and urinary BPA concentrations. In 2009–2011, a convenience sample of 50 adults was recruited from residential settings in North Carolina. The participants completed diaries and collected samples during weeks 1, 2, and/or 6 of a six-week monitoring period. BPA was detected in 38%, 4%, and 99% of the solid food (n = 775), drinking water (n = 50), and surface wipe samples (n = 138), respectively. Total BPA (free plus conjugated) was detected in 98% of the 2477 urine samples. Median urinary BPA levels were 2.07 ng/mL, 2.20 ng/mL-SG, 2.29 ng/mg, and 2.31 ng/min for concentration, SG-adjusted, CR-adjusted, and excretion rate values, respectively. The intraclass correlation coefficient (ICC) estimates for BPA showed poor reproducibility (≤ 0.35) for all urine sample types and methods over a day, week, and six weeks. CR-adjusted bedtime voids collected over six-weeks required the fewest, realistic number of samples (n = 11) to obtain a reliable biomarker estimate (ICC = 0.80). Results of linear mixed-effects models showed that sex, race, season, and CR-level were all significant predictors ($p < 0.05$) of the adults' urinary BPA concentrations. BPA levels in the solid food and surface wipe samples did not contribute significantly to the participants' urinary BPA concentrations. However, a significant positive relationship was observed between solid food intake and urine-based estimates of BPA dose, when aggregated over 24-h periods. Ingestion of BPA via solid food explained only about 20% of the total dose (at the median of the dose distribution), suggesting that these adults were likely exposed to other major unknown (non-dietary) sources of BPA in their everyday environments.

1. Introduction

Bisphenol A (4,4′-dihydroxy-2,2-diphenylpropane, [BPA]), is a synthetic chemical with over two billion pounds produced annually in the United States (US) (Geens et al., 2012; Loganathan and Kannan, 2011). BPA is commonly used to create polycarbonate plastics and epoxy resins for use in everyday consumer products and packaged goods (ACC, 2015). Several studies have found measurable levels of

Abbreviations: BPA, bisphenol-A; BMI, body mass index; CR, creatinine concentration; EPA, US Environmental Protection Agency; Ex-R study, A Pilot Study to Estimate Human Exposures to Pyrethroids using an Exposure Reconstruction Approach; LC/MS/MS, liquid chromatograph - tandem mass spectrometer; HSF, Human Studies Facility; FMV, first-morning void; GC/MSD, gas chromatograph-mass selective detector; GM, geometric mean; ICC, intraclass correlation coefficient; LOQ, limit of quantitation; NHANES, National Health and Nutritional Examination Survey; NC, North Carolina; QC, quality control; QuEChERS, quick, easy, cheap, effective, rugged, and safe method; RTP, Research Triangle Park; SG, specific gravity; SRS, surrogate recovery standard; UPS, United Parcel Service; US, United States

* Corresponding author at: US EPA, 109 T.W. Alexander Dr., MD E205-04, Research Triangle Park, NC 27709, USA.
E-mail address: morgan.marsha@epa.gov (M.K. Morgan).

https://doi.org/10.1016/j.envint.2017.12.014
Received 11 September 2017; Received in revised form 7 December 2017; Accepted 10 December 2017

BPA in many commonly used consumer items including food storage containers, food cans (liners), tableware, paper receipts, electronic equipment, magazines, paints, adhesives, shampoos, bar soaps, body lotions, sunscreens, nail polishes, and recycled paper towels, napkins and toilet papers (Dotson et al., 2012; Geens et al., 2012; Liao and Kannan, 2011; Vandenberg et al., 2007).

Due to its widespread use, BPA has been found in several different types of environmental media including foods, beverages, dust, surface wipes, and air at residences in the US (Loganathan and Kannan, 2011; Rudel et al., 2003; Schecter et al., 2010; Wilson et al., 2007). Although adults can be exposed to BPA though multiple routes, research has indicated that dietary ingestion is likely the dominant exposure route (> 90%) (Geens et al., 2012; Morgan et al., 2011; Von Goetz et al., 2017; Wang et al., 2015). Currently, the US Food and Drug Administration does not regulate the amount of BPA that can be present in foods or beverages for adult consumption (Schecter et al., 2010). Several recent studies have raised concerns that exposures to BPA at environmental concentrations may be adversely impacting human health (i.e., reproductive, developmental, and cardiovascular effects) (Rezg et al., 2014; Rochester, 2013; Schug and Birnbaum, 2014).

Once ingested, BPA undergoes rapid metabolism in the liver and is mainly excreted in the urine as conjugated BPA-glucuronide with an elimination half-life of < 7 h in adults (Dekant and Volkel, 2008; Thayer et al., 2015; Volkel et al., 2002). In recent cross-sectional studies (2009–2014), total BPA was frequently detected (> 73%) in the urine samples of adults recruited from the US general population (CDC, 2017; Cox et al., 2016; LaKind and Naiman, 2015; Ye et al., 2015). In these studies, median urinary BPA concentrations ranged from 0.36–2.4 ng/mL.

A number of US studies have examined the short- or long-term variability (1 week to 3 years) of total BPA concentrations in spot, first-morning void (FMV), and/or 24-h urine samples in adults in non-occupational settings (Braun et al., 2011, 2012; Cox et al., 2016; Meeker et al., 2013; Philippat et al., 2013; Pollack et al., 2016; Reeves et al., 2014; Townsend et al., 2013; Ye et al., 2011). In some of these studies, FMVs has been the preferred sample type collected based on the assumption that they provide the best weighted-average biomarker level over a day (Kissel et al., 2005). None of these prior studies have specifically examined the levels of BPA in bedtime voids. Across these various studies, poor reproducibility (ICC < 0.30) of repeated measurements of BPA occurred in adult urine samples regardless of urine sample type and method of adjustment (unadjusted, specific gravity [SG] adjusted, or creatinine [CR] adjusted). The observed high within-individual variability reported in these studies suggests that a single BPA urine measurement is probably not sufficient to characterize a person's average exposure over a day or longer. However, information is currently lacking on the actual type and number of urine samples that are likely needed to obtain a reliable BPA biomarker estimate for adults over a day or longer.

Several US studies of non-occupationally exposed adults have reported significant ($p < 0.05$) associations occurring between urinary BPA concentrations and various sociodemographic, lifestyle, and dietary factors including age, gender, and race/ethnicity (LaKind and Naiman, 2008, 2015), pre-pregnancy body mass index (BMI) (Meeker et al., 2013), contact with paper receipts (Gerona et al., 2016), and consumption frequency of canned vegetables (Braun et al., 2011), hamburgers (Quiros-Alcala et al., 2013), and soda (LaKind and Naiman, 2008; Quiros-Alcala et al., 2013). Currently, we are not aware of any published study that has quantitatively examined the association between measured BPA levels in the actual consumed diets of adults and their urinary BPA levels.

In previous work from the Pilot Study to Estimate Human Exposures to Pyrethroids using an Exposure Reconstruction Approach (Ex-R study), we quantified the levels of several pyrethroid insecticides and pyrethroid degradates in media (duplicate-diet solid food, drinking water, hard floor surface wipes, and urine) for 50 adults in residential settings over a six-week monitoring period in North Carolina (NC) in 2009–2011 (Clifton et al., 2015; Morgan et al., 2016a; Morgan et al., 2016b; Starr et al., 2017). In this present work, we have now quantified the concentrations of BPA in the same media (above) from the Ex-R study. The major objectives were to: 1) quantify the BPA levels in the duplicate-diet solid food, drinking water, hard floor surface wipe, and urine samples (spot and 24-h) collected from 50 Ex-R adults over a six-week monitoring period; 2) determine the temporal variability of BPA levels in the FMV, bedtime, and 24-h urine samples as concentration, SG-adjusted, CR-adjusted, and excretion rate values, and; 3) examine associations between available study factors and urinary levels of BPA.

2. Materials and methods

2.1. Study cohort

The Ex-R study design and sampling methodology has been described earlier in Morgan et al., 2016a. This study was designed to assess the short-term (over six-weeks) exposures of adults to pyrethroid insecticides and BPA in selected media at residences. Briefly, this observational exposure measurements study was conducted at the US Environmental Protection Agency's (EPA's) Human Studies Facility (HSF) in Chapel Hill, NC, and within a 40-mile radius of the HSF at the adult participants' homes. A total of 50 adults between the ages of 19 and 50 years old were recruited into the study. The participants filled out diaries (food and activity) and collected duplicate-diet solid food, drinking water, hard floor surface wipe, and urine samples (spot and 24-h) during weeks 1, 2, and 6 of a six-week monitoring period from November 2009 to May 2011 (Fig. 1). Each sampling week started on Sunday (day 1) and ended on Friday (day 6). The University of North Carolina's Institutional Review Board approved the Ex-R study protocol and procedures (study number 09–0741) in 2008. All study adults reviewed and signed informed consent forms before participating.

2.2. Collection of diaries and physical information

The Ex-R participants filled out the food and activity diaries during sampling weeks 1, 2, and 6 of the six-week monitoring period (Morgan et al., 2016a). Food diaries were complete on days 1–2 and days 4–5 of each sampling week. Each 24-h sampling day consisted of three consecutive time periods (period 1 = 4:00–11:00 am, period 2 = 11:00 am–5:00 pm, and period 3 = 5:00 pm–4:00 am). The food diary was used to record all of the solid and liquid foods (including estimated amounts) consumed by individual participants during each sampling time period. Activity diaries were completed on days 1–2 and days 4–5 of each sampling week. Each 24-h sampling day in this diary consisted of three consecutive time periods (as described above). The activity diary was used to record the participants' activity levels (sleeping, low [e.g., sitting/standing], medium [e.g., walking], or high [e.g., running]) in consecutive, 30-min time increments during each sampling time period. In addition, an EPA technician recorded specific physical information (weight, height, age, and sex) about each participant at the HSF at the beginning of sampling week 1.

2.3. Collection of samples

The participants were trained to collect their own duplicate-diet solid food, drinking water, hard floor surface wipe, and urine samples during weeks 1, 2, and/or 6 of a six-week monitoring period (Morgan et al., 2016a). Duplicate-diet liquid food samples (mainly beverages, excluding drinking water) were not collected in the Ex-R study because of participant burden and budget constraints. The adults collected duplicate amounts of all solid food samples (e.g., fruits, vegetables, meats, pastas, soups, crackers, cheeses, breads, cookies, and ice creams) they consumed on sampling days 1 and 2 during each sampling week. The solid food samples were collected over three consecutive time periods