Incorporating potable water sources and use habits into surveys that improve surrogate exposure estimates for water contaminants: the case of bisphenol A

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ABSTRACT

Human biomonitoring studies for water contaminants are often accompanied by surveys relying solely on total drinking water consumption rates, thus, failing to account for specific water sources (bottled and tap water) and use habits, such as water used for preparing cold/hot beverages (coffee, tea, juice, etc.). Despite the extensive use of bisphenol A (BPA) in polycarbonate (PC)-based water contact materials, rarely do BPA biomonitoring studies focus on various PC water uses and sources. Better resolved water consumption rates could reduce the uncertainty associated with surrogate daily BPA intake estimates using fine-tuned surveys. This approach provided a proof of concept on inclusion of water consumption from various sources and uses into estimates of daily intake for water contaminants like BPA found in water-contact materials. The next steps would be in quantifying the extent of improvement in exposure assessment that adds value to refined survey designs.

Key words | biomonitoring, bisphenol A, environmental epidemiology, exposure assessment, polycarbonate bottled water, water and health

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INTRODUCTION

In the post-Second World War era, public health officials were already witnessing dramatic decreases in waterborne illness incidence rates in areas utilizing centralized drinking water treatment with pre- and post-chlorination facilities. Since then, research focus on mortality and morbidity risk factors has shifted away from water-based health indicators, while only during the last decade has considerable attention for safe access to water and sanitation issues again been documented for developed countries. Unsafe water, sanitation, and hygiene represent the leading environmental risk factors influencing global mortality and morbidity when compared with other environmental factors, such as urban outdoor air pollution, indoor smoke from solid fuels, environmental lead exposure, and global climate change (Ezzati *et al.* 2002). Water consumption rates are characterized by large inter-population and intra-population variability reflecting differences in age, sex, socioeconomic status, racial characteristics, cultural differences, geographic location, etc. More than a single source of potable water is used in developed countries (such as tap, bottled, desalinated seawater/groundwater, etc.), perplexing estimates of potable water consumption (Makris *et al.* 2013a, b). Water surveys have been traditionally used in epidemiologic, marketing, or economic studies, but they often do not reflect the multitude of water consumption, source and use characteristics for the population of interest.

Bottled water enjoys big sales in many parts of the world, being another popular choice of potable water (Rodwan 2011). However, water packaged into plastic containers could come in contact with leached plasticizers and plastics additives (Andra *et al.* 2011, 2012), such as antimony, bisphenol A, phthalates (to name a few), showing endocrine disrupting activity in key hormonal centers, like reproduction (Wolff *et al.* 2008a) and thyroid (Andra & Makris 2012). Exposure assessment of water contaminants,

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including those in bottled water, has not gained as much attention as food contaminants enjoy in major pregnancy–birth cohorts (Makris & Andra 2014). With the exception of disinfection by-products, potable water contaminants like endocrine-disrupting chemicals have received little, if any, attention in biologically relevant exposure assessment for environmental epidemiological studies (Makris & Andra 2014). Exposure assessment for water contaminants has typically relied upon water intake survey questionnaire information (Smith *et al.* 2009; Llop *et al.* 2011; Haugen *et al.* 2011) rather than water chemical analysis (Makris & Andra 2014). Where available, water intake survey questionnaires tend to rely on contemporary questions such as 'how many glasses of water do you daily consume?', which do not provide explicit details that could better drive estimates of daily water intake for water pollutants such as disinfection by-products (Maskiell *et al.* 2006) and perchlorate (Mendez *et al.* 2010).

The example of bisphenol A (BPA) is exemplified here, since it is a controversial endocrine disruptor found in polycarbonate (PC)-based water contact materials (WCM), among other sources (cans, baby bottles, personal care products, etc.) (Von Goetz *et al.* 2010; Makris *et al.* 2013b; Muncke *et al.* 2013). Typically, the source of BPA occurrence in tap water is epoxy resin coatings or polyvinyl chloride material used in drinking water distribution system pipes (Bae *et al.* 2002). However, BPA is not detected or present at very low concentrations in tap water (Makris & Snyder 2010) because of chlorine-induced degradation of BPA to chlorinated BPA congeners (Hu *et al.* 2002). However, BPA in packaged water may be expected to be measured, since chlorination of packaged water is not practiced unless a previously chlorinated potable water source was used for packaging. Traditional survey questions relying on total water consumption per capita often fail to address specific water source(s), use and frequency characteristics (cups/glasses per day or week), poorly reflecting upon biologically relevant exposure estimates of water contaminants. Specific water uses refer not only to water used for plain drinking water purposes, but also water used for making cold (frappe, coffee, juice, etc.) and/or hot (tea, coffee, etc.) beverages, or water used for cooking. It is often subtle differences and differentiation of water sources (various plastic types and volumes of bottled water, well water, mobile stalls selling mountain water, etc.) and water uses in surveys that could link biomonitoring-based exposure estimates to external water source(s). As an example, PC-based water consumption, rather than total (both bottled and tap) water consumption was significantly ($p = 0.017$) associated with urinary BPA concentrations in a small ($n = 35$) young adult subpopulation group that largely relied upon PC bottled water to satisfy its potable needs (Makris *et al.* 2013b). Polyethylene terephthalate (PET)-based water consumption patterns showed a significant ($p = 0.02$) positive correlation with urinary antimony concentrations, linking the antimony-containing PET bottled water to human internal exposures (Makris *et al.* 2013a). When total water consumption patterns were used (data derived from survey-based questions like 'how many glasses of water per day you consume?'), no correlation was observed with either antimony or BPA (Makris *et al.* 2013a, b).

Given the distinct differences and gaps in surveys used in epidemiological studies monitoring endocrine disruptors' exposures from various environmental stressors, including water contamination, we aimed at further improving exposure assessment with the suggestion of a fine-tuned questionnaire probing into individual preferences of water source (s) and use characteristics. Despite the small size of our study ($n = 35$), it successfully showed the effect of refining water-related surveys for environmental contaminants onto improving our ability to associate specific exposure sources to body burden estimates. The superiority of fine-tuned survey content over that of a contemporary survey was proven within the same pilot study of ours. Though outcomes of this fine-tuned survey questionnaire as a relationship between per capita water consumption and daily intake of either antimony from PET bottled water (Makris *et al.* 2013a) or BPA from PC bottled water (Makris *et al.* 2013b) was earlier reported, the novelty of this submission is in presenting the superiority and differences in intake estimates (equivalent to exposure) compared with a contemporary approach used in parallel, considering urinary BPA data as a case study. We also searched the literature to assess the extent of water (either tap or bottled) contaminant studies following such refined water-related surveys. The overall objective was to emphasize the need for detailed survey questions while assessing exposures to endocrine disruptors found in food and water-contact materials, like BPA.

METHODS

In an effort to improve the environmental exposure estimates explaining the variability in chronic disease incidence rates, a refined survey content emphasizing hypothesis-driven exposure sources and pathways was applied in an exposure assessment study for BPA (Makris *et al.* 2013b). In comparison, we gathered human biomonitoring-based studies from a literature search engine that reported geometric means of either creatinine-corrected or unadjusted urinary BPA levels. A comprehensive search was performed using Scopus queried from 1960 (start date) to March 1, 2013. The keyword search 'bisphenol AND water' yielded 3,200 peer-reviewed publications, which narrowed down with specific keyword combinations such as 'bisphenol AND water AND exposure' resulted in 400 articles. About 200 articles relevant to the environmental sciences were selected, and both abstract and full length text (where necessary) were read and assessed carefully in summarizing the available studies. Thirty-six studies were identified reporting urinary BPA in an exposure and/or epidemiological study perspective. The majority of biomonitoring studies reported urinary BPA concentrations as geometric mean and they were retained for further analysis (28 out of 36 available studies, Table SI-1, available online at http://www.iwaponline.com/wh/012/068.pdf). Only a few studies reported arithmetic mean and median values of urinary BPA, three (Kim *et al.* 2003; Tsukioka *et al.* 2003; Yang *et al.* 2003) and five studies (Yang *et al.* 2006; Schoringhumer & Cichna-Markl 2007; Wolff *et al.* 2008b; Meeker *et al.* 2010; Casas *et al.* 2011), respectively. These were excluded from further consideration to facilitate data comparison across all studies reporting urinary BPA in the same statistical expressions (geometric mean and geometric standard deviation). Following this effort, we evaluated how well the refined survey content matched that presented in the selected BPA studies relying on a contemporary survey content which entailed a single question of 'how much water do you consume per day' (Table SI-1).

Survey study and participants

Study details are presented elsewhere (Makris *et al.* 2013a). In brief, a pilot study of 35 adult volunteers (university post-graduate students, faculty, and staff) was conducted to identify relations between water intake pattern and associated exposures to endocrine disrupting compounds. A detailed fine-tuned questionnaire was administered and compared the outcomes with those of a contemporary survey approach (detailed below). Urinary antimony and BPA were used as a measure of water consumption from PET and PC bottles, respectively (Makris *et al.* 2013a, b). The mean [±SD (standard deviation)] age and body mass index (BMI) of the study participants was 30.5 ± 7.5 years and $23.6 \pm 4.3\ \text{kg m}^{-2}$, respectively. The majority were females (63%) in the age group 26–35 years (57%), with a normal BMI of $<25\ \text{kg m}^{-2}$ (74%).

Survey approach

Both contemporary and fine-tuned survey questionnaires were implemented in parallel (provided together and at the same time). Graphical representation of the steps involved in these approaches is presented in Figure 1. Details are provided below as purposes and approaches.

Purpose-I

Determine the type and extent of use of a water source to meet daily potable water consumption requirements.

- Pointers: Pictures of each water source should be shown to participants towards better addressing this question. A 250 mL glass or cup needs to be displayed as an indicator for cup volume water consumption. If the participant provides answer for the 'other' source, the interviewer should make a special note of it for further follow up.
- Posed questions:
  - Q1: Which of the following one or more sources of water do you use – a YES or NO response for: (1) water board of city (tap water), (2) ground water (well water), (3) bottled water, (4) mobile stations (water stalls on the road filled with mountainous spring water), (5) other, please specify.
  - Q2: If YES, please specify how many glasses (a glass equals 250 mL) of water per day and how many days per week from each water source.
- Possible outcome: Provides an overall depiction of major water sources and an estimate of water intake from each

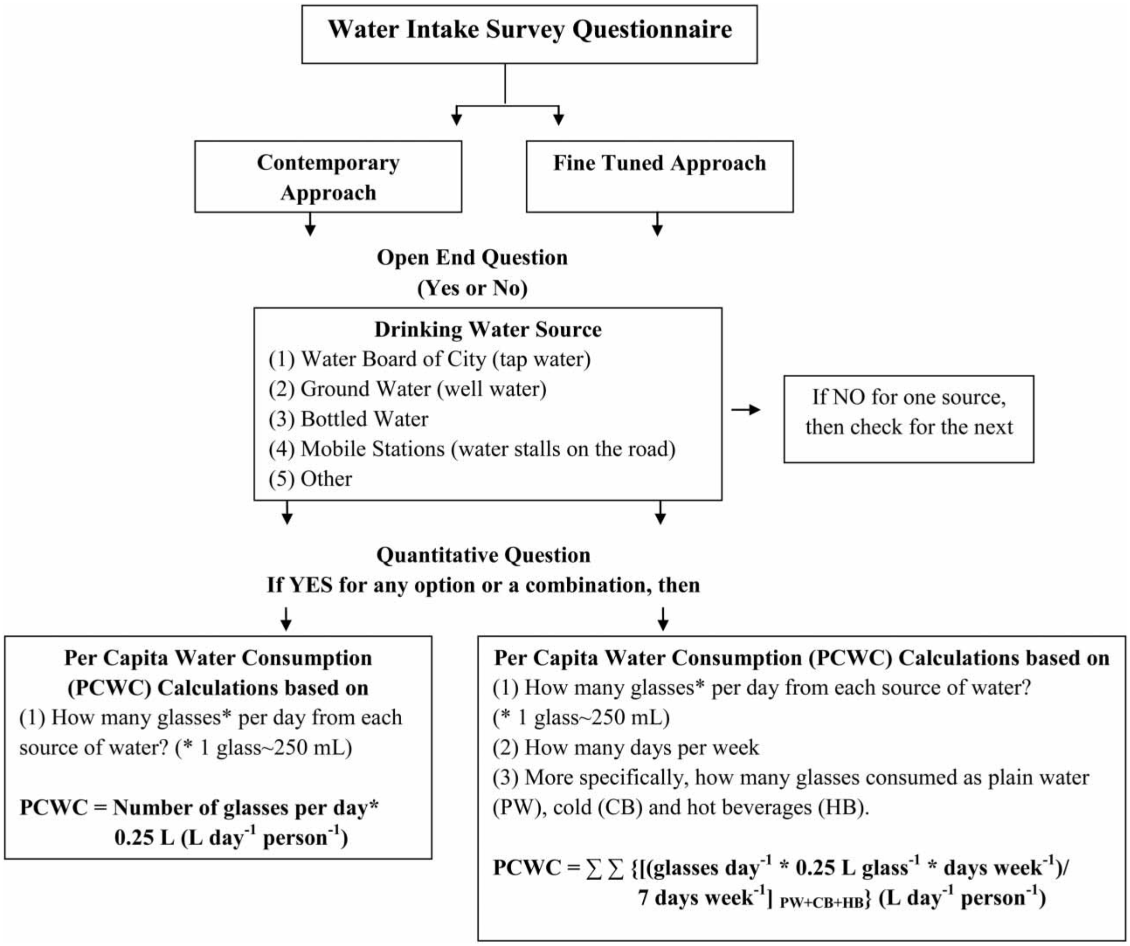

Figure 1 | Steps followed in assessing water consumption via contemporary and fine-tuned approach.

source. This leads to further understanding on the extent of use of water sources 'other' than tap water, which is usually considered as the predominant, and sometimes as the one and only, source of water.

Purpose-II

To determine which plastic type of bottled water was used.

- Pointers: A brief on types of plastics used in consumer products and examples of daily sources of exposure to plastics should be mentioned to each participant. It is necessary to carry a schematic of plastics codes from 1 through 7, corresponding details about each resin identification code, and a mention on location of the code on plastic containers. A brief note on plastics additives chemical nature and associated human health perturbations is recommended. PET bottles are coded 1 and widely used because of their convenience to carry, but recommended for one time use. PET material likely contains phthalates (known endocrine disrupting chemicals, EDC) and antimony (possible EDC and a carcinogen). PC bottles are coded 7 and widely used as large water storage containers that are usually reused. PC material likely contains BPA, a known endocrine disruptor. PET and PC material is used not only for bottling water but also for packaged beverages and food. Pictures of both PC (19 L) and PET (0.5/0.75/1.5 L) must be shown to the participants for a better response to the questionnaire. If a participant provides an unrealistic number of glasses of water consumption per day, the interviewer must specifically seek the correct answers for 'how many days per week from each bottle type' – since preferences may change from while at home and when being outside home (e.g., at work).

- Posed questions:
  - Q1: Which one of the following plastic bottles did you use for water consumption – a YES or NO response for: (1) polyethylene terephthalate (PET), (2) polycarbonate (PC), (3) both, please specify.
  - Q2: If YES for PET, please specify the bottle volume – a YES or NO response for: (1) 0.5 L, (2) 0.75 L, (3) 1.5 L, (4) all.
  - Q3: Where and when a YES response from Q1 and Q2, please specify how many glasses (250 mL) of water per day and how many days per week from each plastic bottle type.
- Possible outcome: Provides a detailed description on bottled water type preference in the survey population. This furthers helps in identifying possible exposures to respective endocrine disruptors that vary with plastic type – for example phthalates and antimony used in PET bottles and BPA in PC bottles. Moreover, the effort from this step increases the awareness of study participants on various plastics used in consumer products, associated health concerns, and recycling capabilities of each type.

Purpose-III

To determine the use of water for which other consumption purposes other than plain drinking water.

- Pointers: Pictures of cold and hot beverages may help in making better sense of posed questions by the participants. Again, if the interviewer gets an unrealistic number from the participant, then it is recommended to re-pose the questions for a better breakdown of water consumption pattern towards cold and hot beverages.
- Posed questions:
  - Q1: Which one of the following purposes did you use water for consumption – a YES or NO response for: (1) plain drinking water, (2) making cold beverages (juice, mixed drink, etc.), (3) making hot beverages (tea, coffee, etc.), and (4) all of the above or a combination, please specify.
  - Q2: If YES, please specify how many glasses (250 mL) of water per day and how many days per week from each water source.
- Possible outcome: Describes the distribution of water source usage among daily water use types. It also presents information on some of the neglected water use purposes in a generic survey questionnaire.

Calculations

Per capita water consumption using the fine-tuned approach was calculated by using the formulae:

$$\text{Daily water consumption rates, L day}^{-1}\text{person}^{-1} = \sum\left[X_{\text{Plain Water}} + X_{\text{Cold Beverages}} + X_{\text{Hot Beverages}}\right] \quad (1)$$

$$\text{where } X = \left(\text{glasses day}^{-1} * 0.25\ \text{L glass}^{-1} * \text{days week}^{-1}\right) / 7\ \text{days week}^{-1} \quad (2)$$

Urinary BPA analysis

Urinary BPA (total of free and conjugated BPA) was analyzed using a liquid chromatography–mass spectrometry method. The experimental and analytical details of the method are detailed elsewhere (Makris *et al.* 2013b). The primary focus of this work was to highlight the differences in daily intake estimates derived from using both contemporary and a fine-tuned survey questionnaire on per capita water consumption rate and BPA intake (back calculated from urinary BPA). Descriptive statistics, univariate and correlation analyses were performed using JMP 7.0 (SAS Inc., NC, USA).

RESULTS AND DISCUSSION

Compiling data from 28 human biomonitoring studies that reported geometric mean BPA concentrations helped us evaluate whether details about water source and usage characteristics were taken into consideration when estimating daily intake estimates (Table SI-1, available online at http://www.iwaponline.com/wh/012/068.pdf). With respect to water consumption per source as a question, bottled water was surveyed in the studies by Yang *et al.* (2006), Carwile *et al.* (2009), Lakind & Naiman (2011), Morgan *et al.* (2011), and Li *et al.* (2013), and tap water was studied by Völkel *et al.* (2008), Becker *et al.* (2009), and Li *et al.* (2013). Makris *et al.* (2013b) included other water sources, such as ground water and mobile stations which is water packaged in reusable PC containers. Regarding container type, PC bottled water consumption was surveyed by Carwile *et al.* (2009) and Li *et al.* (2013). In addition to PC bottles, water consumption from PET bottles was assessed by Makris *et al.* (2013b). With respect to water use characteristics, consumption in the form of plain water was taken into consideration by Völkel *et al.* (2008), Carwile *et al.* (2009), and Li *et al.* (2013), while in the form of cold beverage by Carwile *et al.* (2009) and hot beverage by Li *et al.* (2013). All three forms of water use (plain water, and cold and hot beverages) were monitored by Makris *et al.* (2013b). Though urinary BPA levels were measured in the 28 studies, neither water consumption source nor water use specifics were reported by 20 out of the 28 studies listed in Table SI-1 (viz., Yang *et al.* (2003), Calafat *et al.* (2005), Wolff *et al.* (2007), Calafat *et al.* (2008), Lakind & Naiman (2008), Mahalingaiah *et al.* (2008), Teitelbaum *et al.* (2008), Wolff *et al.* (2008b), Ye *et al.* (2008), He *et al.* (2009), Bushnik *et al.* (2010), Cantonwine *et al.* (2010), Galloway *et al.* (2010), Mendiola *et al.* (2010), Milieu en Gezondheid (2010), Mok-Lin *et al.* (2010), Braun *et al.* (2011), Kim *et al.* (2011), Zhang *et al.* (2011), and Nahar *et al.* (2012)). Among the 28 BPA studies available, only three studies included more than one single water source in their questionnaire, because often consumers in developed countries use two or more water sources for potable use (Carwile *et al*. 2009; Li *et al*. 2013, Makris *et al.* 2013b) (Table SI-1). These three studies were those that included questionnaire items looking into water use characteristics (water used for cold or hot beverages) (Table SI-1). Indeed, most of the studies in Table SI-1 did not consider the possible contribution of any water source and/or use characteristics to daily BPA intake estimates back-calculated from urinary biomarker measurements.

About 50% higher per capita water consumption was reported using the proposed fine-tuned approach (the breakdown resulted in a mean water consumption estimate of 3.44 L day^{-1} $person^{-1}$, while a contemporary approach indicated a mean water consumption of about 1.85 L day^{-1} $person^{-1}$; Table 1). Use of plain drinking water topped the potable water consumption chart, while a significant proportion (30–35% of total daily water consumption) took the form of cold and/or hot beverages (coffee, tea, juices). The observation became apparent because this study was conducted during the summer period of higher water consumption and use (Table 1). The refined questionnaire indicated that 35% of per capita daily water consumption would go unreported, if only plain drinking water was taken into account (Table 1). The 'use of other water source' question did not yield any answers, while the intention was to seek information on alternative water sources, such as surface water (lake, etc.). This indicates the need for going over, one by one, all water source options rather than having the participant think through them.

Significant association ($p = 0.02$) was observed between water consumption from PC bottles derived from the fine-tuned approach and urinary BPA (Figure 2(d)). No such significance ($p > 0.05$) was noted between urinary BPA and water consumed from all sources (Figure 2(a)), all bottled water (Figure 2(b)), and water from both PC and PET bottles (Figure 2(c)). The ability to take into account the total use of PC bottled water in a systematic manner using a fine-tuned approach helped in finding a meaningful association between PC bottled water consumption and BPA intake (measured as urinary BPA). Otherwise, the estimates using a contemporary approach yielded no such significant association with BPA intake due to an inherent deficiency of failing to consider the contribution of PC bottled water towards per capita water consumption. Furthermore, no such significant linear trends ($p > 0.05$) were observed between urinary BPA levels and water consumption from PET bottles (Figure SI-1b) and tap (Figure SI-1c) that were also derived from the fine-tuned survey data. (Figure SI-1 is available online at http://www.iwaponline.com/wh/012/068.pdf.)

Most of the existing water use surveys in search of associations with urinary BPA relied on either daily water consumption or an average of two consecutive days as in the United States National Health and Nutrition Examination Survey (NHANES) (Lakind & Naiman 2008; Sebastian *et al*. 2011). However, the fine-tuned approach in this study showed that relying on water consumption information for a day or two may not be a good representation given the variable number of hours spent weekly by individuals at work, leisure time and at home. For example,

Table 1 | Development of a fine-tuned questionnaire with respect to per capita water consumption data (L day^{-1} $person^{-1}$) in comparison to a contemporary approach, for water contaminants that often show multiple exposure sources. Values represent mean ± standard deviation of 35 adult volunteers

		Average number of glasses day^{-1} (= glasses day^{-1} * days $week^{-1}$/7 day $week^{-1}$)	Per capita water consumption (L day^{-1} $person^{-1}$)
Contemporary approach (with no additional classifications in comparison to fine-tuned approach)			
Bottled water		3 ± 3	0.86 ± 0.80
Tap water		3 ± 4	0.65 ± 1.05
Ground water		0 ± 1	0.06 ± 0.28
Mobile station water		1 ± 4	0.27 ± 0.90
Contemporary approach: Cumulative per capita water consumption		**7 ± 5**	**1.85 ± 1.18**
Fine-tuned approach (further classification based on water source, use, and plastic type)			
Bottled water			
Polycarbonate (PC 19 L/big blue container)	Plain water	3 ± 2	0.63 ± 0.61
	Cold beverage	1 ± 1	0.20 ± 0.27
	Hot beverage	1 ± 1	0.18 ± 0.26
Total PC water		*4 ± 3*	*1.02 ± 0.79*
Polyethylene terephthalate (PET 0.5/0.75/1.5 L)	Plain water	3 ± 4	0.74 ± 0.89
	Cold beverage	1 ± 2	0.21 ± 0.40
	Hot beverage	1 ± 1	0.13 ± 0.24
Total PET water		*4 ± 5*	*1.08 ± 1.24*
Total bottled water		*8 ± 6*	*2.10 ± 1.42*
Tap water	Plain water	3 ± 4	0.74 ± 0.89
	Cold beverage	1 ± 1	0.13 ± 0.25
	Hot beverage	1 ± 1	0.15 ± 0.22
Total tap water		*3 ± 5*	*0.81 ± 1.29*
Ground water	Plain water	0.20 ± 0.9	0.05 ± 0.23
	Cold beverage	0.02 ± 0.1	0.01 ± 0.03
	Hot beverage	0.02 ± 0.1	0.01 ± 0.04
Total ground water		*0.24 ± 1.0*	*0.06 ± 0.26*
Mobile station water	Plain water	1 ± 3	0.26 ± 0.71
	Cold beverage	1 ± 3	0.17 ± 0.64
	Hot beverage	0.2 ± 0.7	0.04 ± 0.18
Total mobile station water		*2 ± 6*	*0.48 ± 1.43*
Fine-tuned approach: Cumulative per capita water consumption		**14 ± 8**	**3.44 ± 2.02**

participants usually have a wider choice of water sources and also water use needs when at work or in school on workdays, while the options may differ on a weekend. Hence, we took a 'frequency approach' to average out water consumption over a 1-week period by integrating responses from two survey questions: (1) how many glasses (250 mL) per day from each water source, and (2) how many days per week from each water source? As noted, there was an inflation in mean water consumption and a huge standard deviation when calculations were based on a daily basis in comparison with frequency (weekly) basis (Figure SI-2, available online at http://www.iwaponline.com/wh/012/068.pdf). A fine-tuned approach resulted in an average (±SD) per capita water consumption of 5.0 ± 3.1 and 3.4 ± 2.0 L day^{-1} $person^{-1}$ from all water sources using daily and weekly based calculations, respectively (Figure SI-2). Similarly, the per capita water

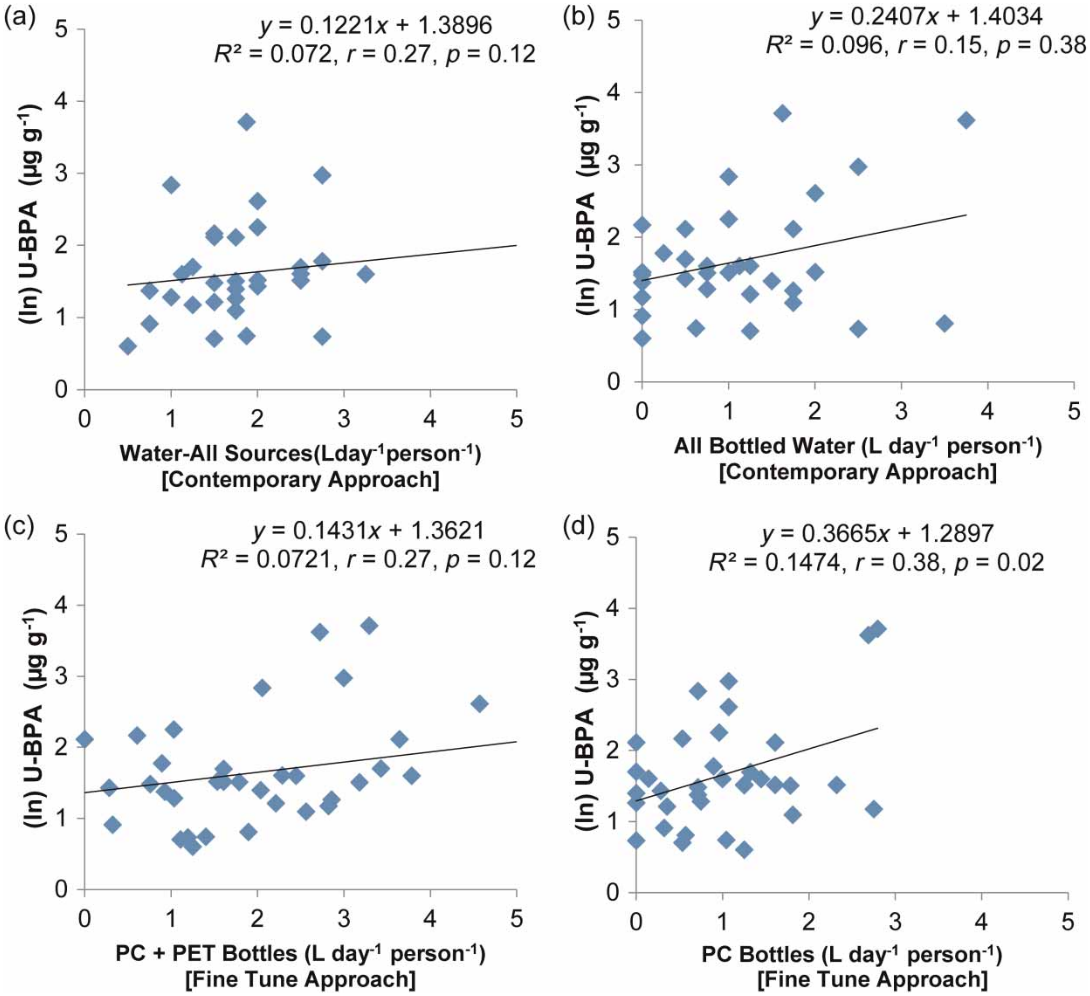

Figure 2 | Creatinine-adjusted BPA concentrations in the survey study participants ($n = 35$) in relation to the reported per capita water consumption from all drinking sources (a) and only bottled water (b) using a contemporary survey. Similar relations were graphed between urinary BPA levels and per capita water consumption from the refined PC + PET water survey (c) and only PC bottled water (d).

consumption from only plastic bottles (all bottled water) using a daily and weekly based calculation was 3.2 ± 2.1 and 2.1 ± 1.4 L day^{-1} person^{-1}, respectively (Figure SI-2). On the other hand, the PC water consumption estimate based on plain drinking water purpose alone was 0.6 ± 0.6 L day^{-1} person^{-1}, which significantly increased to 1.0 ± 0.8 L day^{-1} person^{-1} upon inclusion of PC water use for cold and hot beverages ($p = 0.03$) (Figure 3). The resulting increased contribution of PC water consumption from inclusion of other water use aspects significantly enhanced the BPA intake estimate from 24 ± 24 to 40 ± 32 ng kg bw^{-1} day^{-1} ($p = 0.04$) (Figure 3). This observation further supports the deemed necessity in considering alternative uses of PC water that may significantly increase both per capita water consumption and BPA intake estimates, which has gone unnoticed and under-considered thus far in conducting risk assessments.

The proposed fine-tuned approach, however, suffers from limitations, because the provision of a detailed questionnaire might induce a redundant tendency to overestimate water consumption rates from each source and use, thereby overestimating water-based BPA intake rates. The other limitation would be lack of a 'gold standard' in drinking water survey content to compare and/or validate our study questionnaire with. Similarly, higher rates of water consumption and greater variation were reported during

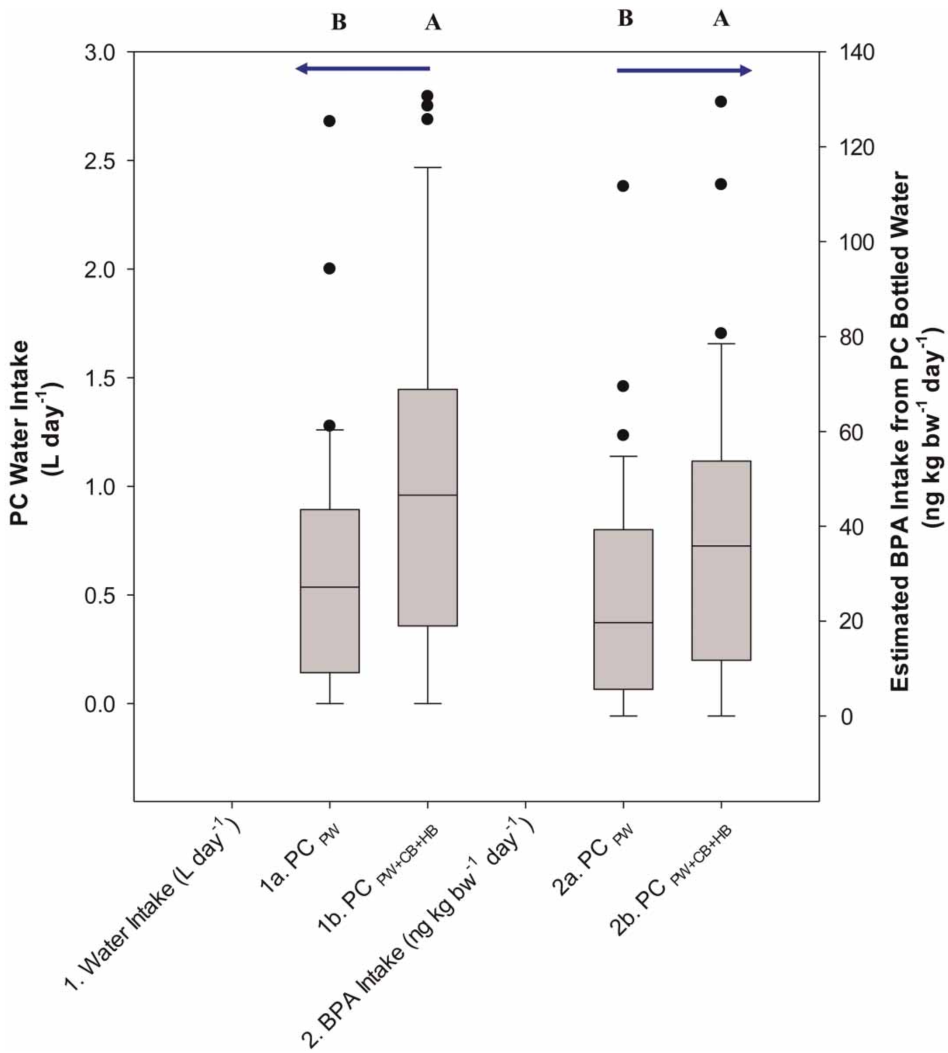

Figure 3 | Water use characteristics (either as plain water or beverage or both) gives a closer estimate on per capita water consumption from PC bottles and associated BPA intake by the 35 adult volunteers participating in this study. Mean values denoted by different letters are statistically significant using Student's t-test ($\alpha = 0.05$).

summer and winter periods, with an average total per capita water consumption of 38.6 and 34.6 oz day^{-1} person^{-1} (1.14 and 1.02 L day^{-1} person^{-1}), respectively, and ranged from 0 to 396 oz (0 to 12 L day^{-1} person^{-1}) (Barraj *et al.* 2009). An additional limitation would be the small sample size ($n = 35$) of the study, preventing us from making inferences for the general population. Hence, the results should be interpreted with a certain degree of caution. Nonetheless, given that there is very little information available on (1) usage of more than one potable water source and (2) usage for more than one water use, this critical examination of questionnaire readouts and the initial descriptive analyses are a useful first step and contribution to the literature. The potential strength of this study was that the results indicate new areas of research interest in deriving finer exposure estimates to drinking water contaminants. The findings of this validation study are of the utmost importance as a proof-of-pilot-concept for further exploration and exploitation. Optimization, validation, and rigorous testing of the proposed questionnaire are the necessary next steps warranted for future application in survey-based health studies.

Adult human exposures to BPA stem primarily from dietary sources such as canned food (>90% contribution) (Geens *et al.* 2010); while the contribution of non-dietary sources is much smaller, such as dust (<1%) (Loganathan

& Kannan 2011), thermal and other paper (<5%) (Liao & Kannan 2011a), currency paper (<1%) (Liao & Kannan 2011b), and dental surgery (<5%) (Von Goetz *et al.* 2010). Within dietary sources, PC bottled water as an exposure source has received little attention compared to canned food and beverages, perhaps because BPA leaching is not instantaneous but may be the combined kinetic influence of various environmental factors like UV, temperature, re-use, etc. Hence, the development of a fine-tuned questionnaire could help us in reducing uncertainty associated with water consumption, water intake and individual risk estimates for pollutants found in water, but also in other exposure sources (diet, air, soil, dust). The far-reaching aim will be the development of similar questionnaires for emerging chemicals found in consumer products for which biomonitoring studies are yet unavailable, such as UV filters that have shown disruption of both reproductive (Weisbrod *et al.* 2007) and thyroid function in animal studies (Schmutzler *et al.* 2007).

CONCLUSIONS

Human biomonitoring studies for water contaminants are often accompanied by surveys relying solely on total drinking water consumption rates, thus failing to account for specific water sources (bottled and tap water) and use habits, such as water used for preparing cold/hot (coffee, tea, juice, etc.) beverages. Despite the extensive use of BPA in PC-based WCM, rarely do BPA biomonitoring studies focus on various PC water uses and sources. For the first time, this study presents an approach to fine-tune water consumption estimates forming an integral part of exposure assessment to unintentional intake of dietary contaminants, such as BPA. Taking a detailed approach with different water use fraction categories in the survey helped in refining per capita water consumption calculations, which further improved our ability to target relevant exposure sources. Better resolved water consumption rates could reduce the uncertainty associated with surrogate daily BPA intake estimates using fine-tuned surveys.

This approach could help not only in deriving realistic exposure estimates for contaminants from drinking water, particularly in a cohort study, but also assist the regulatory and industrial agencies with risk assessment. The next steps would be to quantify the extent of improvement in exposure assessment of epidemiological studies that adds value to the proposed survey design. Eventually, this exercise could help identify specific sources and routes of exposure, detailing the extent of exposure contribution from each source.

The next phase of this work is in progress, where approximately 400 residents from 200 households are participating in a cross-sectional study in Cyprus. One of the major objectives was to find associations between detailed water consumption patterns (tap and bottled water per capita consumption patterns as identified in this study) and unintentional human exposures to a suite of drinking water contaminants (measured as urinary levels of disinfection by-products and bisphenol A). The aforementioned study will help us in providing a wide range of individual level data on age, indicators of socio-economic, habitual, and lifestyle factors, and medical history. Thus, this facilitates the opportunity to practice the approaches presented in this study at a much larger scale in a general population with no selection bias.

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