Water Research 99 (2016) 1–6

Contents lists available at ScienceDirect

Water Research

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

Measuring water ingestion from spray exposures

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ARTICLE INFO

Article history:
Received 5 January 2016
Received in revised form
25 March 2016
Accepted 14 April 2016
Available online 20 April 2016

Keywords:
Recycled water
Water reuse
Sprays
Exposure assessment
Cyanuric acid

ABSTRACT

Characterisation of exposure levels is an essential requirement of health risk assessment; however for water exposures other than drinking, few quantitative exposure data exist. Thus, regulatory agencies must use estimates to formulate policy on treatment requirements for non-potable recycled water. We adapted the use of the swimming pool chemical cyanuric acid as a tracer of recreational water ingestion to permit detection of small water volumes inadvertently ingested from spray exposures. By using solutions of 700–1000 mg/L cyanuric acid in an experimental spray exposure scenario, we were able to quantify inadvertent water ingestion in almost 70% of participants undertaking a 10 min car wash activity using a high pressure spray device. Skin absorption was demonstrated to be negligible under the experimental conditions, and the measured ingestion volumes ranged from 0.06 to 3.79 mL. This method could be applied to a range of non-potable water use activities to generate exposure data for risk assessment processes. The availability of such empirical measurements will provide greater assurance to regulatory agencies and industry that potential health risks from exposure to non-potable water supplies are well understood and adequately managed to protect public health.

1. Introduction

Characterisation of exposure levels is a fundamental requirement of health risk assessment; however, for water exposures other than drinking, quantitative exposure data are sparse. In the absence of empirical data, some health agencies have used estimates of inadvertent water ingestion for a range of common exposures (e.g. garden watering, toilet flushing) in order to formulate standards and guidelines for non-potable recycled water made from sewage effluent or stormwater (NHMRC/EPHC/AHMC, 2006). Methods to experimentally document water ingestion for non-potable water uses would provide more certainty for regulatory authorities, and potentially allow comparison of different exposure scenarios and testing of exposure reduction measures.

Ingestion of water during swimming has been successfully measured using cyanuric acid (CYA, CAS No 108-80-5); a chemical which is added to outdoor pools to prevent decomposition of the chlorine disinfectant by ultraviolet light (APSP, 2011). When ingested, CYA is rapidly absorbed, then excreted in the urine without being metabolized (Hammond et al., 1986). Assuming urinary excretion of 100% within 24 h (Allen et al., 1982), an assessment of pool water ingestion can be made by measuring the CYA concentration in pool water and in urine collected over the 24 h period immediately following a swimming event in an outdoor pool (Dorevitch et al., 2011a, 2011b; Dufour et al., 2006).

The objective of the present study was to test whether this methodology could be adapted to measure inadvertent water ingestion from spray exposure in a simulated car wash scenario. We also assessed dermal absorption of CYA to ensure this was not a significant contributor to the measured exposures.

2. Methods

2.1. Study participants

Ethical approvals for the research were obtained from the Human Research Ethics Committees of RMIT University (Project numbers 44/12 and 14/13) and Monash University (Project numbers 2012001779 and CF13/1431–2013000748). Participants aged 18 years or older who did not report significant skin or eye irritation associated with swimming pools or a medical history of kidney disease were recruited from among staff and students of RMIT and Monash Universities.

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http://dx.doi.org/10.1016/j.watres.2016.04.034
0043-1354/

2.2. CYA solution for spray exposure and skin absorption tests

The CYA solution for skin absorption tests and spray exposure tests was made at a concentration of 1000 mg/L after preliminary sensory effects tests showed this to be acceptable to participants. Details of sensory effects tests and preparation of CYA solution are provided in the Supplementary Material.

2.3. CYA assays

The analytical method for measuring CYA in water and urine involved: (a) three stage solid phase (SP) extraction to remove organics and inorganic salts which may interfere with the analysis of CYA (b) derivatisation of CYA to reduce polarity and increase the volatility of CYA and (c) GC/MS analysis (Sinclair et al., 2016). Method validation was performed as per the National Association of Testing Authorities, Australia procedure (NATA, 2009). CYA standards were prepared using analytical grade chemical in a blank urine matrix (urine from anonymous donors subjected to SP extraction to remove contaminants). The resultant standard curve was linear over the range 0.05–1.0 mg/L ($R^2 = 0.997$). The Limit of Detection (LoD) for CYA in urine samples was 0.01 mg/L and the Limit of Quantitation (LoQ) was 0.05 mg/L. SP extracts were shown to be stable on storage at 4 °C for at least 21 days.

Urine samples returned by participants were stored at 4 °C for a maximum of 24 h before SP extraction. SP extracts were analysed immediately by GC/MS or stored at 4 °C for up to 3 days before assay. Following measurement of CYA concentrations in urine samples, the total amount of CYA contained in the 24 h urine collection was calculated from the urine volume. For spray exposure experiments, the volume of CYA solution ingested by the participant was calculated from the CYA concentration measured in the solution to which they were exposed, using an assumption of 100% excretion.

2.4. General procedures for participants

Participants were asked not to swim in a swimming pool in the 48 h prior to the test and during the 24 h urine collection period. They were asked to visit the bathroom to empty their bladder immediately prior to undertaking the test, and after the test procedure they were supplied with urine collection equipment and asked to collect all urine produced during the following 24 h. They were instructed to keep the urine away from direct heat sources but were not required to refrigerate it. Fluid intake was not restricted or monitored during the collection period. The volume of urine returned by each participant was calculated by weight, assuming a specific density of 1.0 g/1 mL. Participants were asked whether they had spilled or forgotten to collect any urine, and if so, to estimate the approximate volume and time of loss.

2.5. Skin absorption tests

For the skin absorption tests, five participants sat with both forearms in a horizontal position and immersed in CYA solution (1000 mg/L) to a depth of about 2 cm above the elbow joint for a period of 10 min. The CYA containers (one for each arm) were covered with lids to prevent splashes. At the end of the immersion period, the forearms were rinsed under running tap water and patted dry with paper towel. All urine produced for the next 24 h was collected and analysed for CYA.

To calculate the volume that would have been absorbed through the smaller exposed area (part of the face) in the spray exposure tests, we used information from the US Environmental Protection Agency on adult male body surface area for chemical exposure assessment (EPA, 2011). According to this source the average percentage of total body area for the relevant body parts are head 6.6% (excluding neck), hands 5.2% and arms (excluding hands) 15.2%. We estimated that the exposed area in the skin absorption tests was 14.3% in total, comprising 9.1% for the forearms (estimated as 60% of the total arm surface area) and 5.2% for the hands. For the spray exposure tests the exposed area was estimated at 1.65% of the total body area (25% of the head). Thus it would be expected that CYA absorption during the spray exposure tests would be about 0.12 of that experienced during the skin absorption tests (ratio of exposed areas 1.65%/14.3% = 0.12).

2.6. Spray exposure tests

A fibreglass half-car replica was constructed and assembled on a wooden frame in the laboratory. The replica was fitted with wheel rims, tyres, side windows, full front and back windscreens and side mirror (as shown in Supplementary Material Fig. S1) to provide a representative experience of car washing. Participants wore a protective coverall with hood, vinyl gloves, waterproof footwear and safety glasses. They carried out a simulated car wash activity for 10 min using a domestic model high pressure spray device (measured flow rate 5.9 L/min, maximum rated pressure 1740 psi/2012 MPa) drawing CYA solution (700–1000 mg/L) from the storage vessel. A research assistant observed the activity and recorded the percentage of time each participant had their mouth open and whether their face was visibly wet at the end of the 10 min. Participants were also asked whether they thought they had swallowed any water (possible responses: none, a few drops, a teaspoon, a mouthful or more). Participants then removed the protective clothing and were provided facilities to wash their face with tap water. All urine produced for the next 24 h was collected and analysed for CYA. A sample of the CYA solution from the storage vessel was also analysed. For seven of the participants, a pre-exposure urine sample was collected and analysed to verify the absence of CYA in urine prior to the exposure test.

2.7. Modeling of spray exposure

Prior to undertaking this spray exposure study, we carried out preliminary tests to verify the reported 100% excretion of CYA within 24 h (Allen et al., 1982), but observed variability in the excretion of a known CYA dose (Sinclair et al., 2016). Our tests on 26 subjects showed the mean excretion of a 1.0 mg ingested dose was 85.3% (95% confidence interval 77.4%–93.2%). In order to account for the effect of this variation during the spray exposure study, we used Monte Carlo simulation to estimate the adjusted ingestion volume using data from the known dose excretion tests (Sinclair et al., 2016). Distribution fitting of the known dose data set was conducted using @Risk Version 5.7.0 (2010, Palisade Corporation, Ithaca, New York). A triangular distribution (minimum 29.3, mode 105, maximum 105) produced the best fit for the data (Chi-Sq. 5.585). The ingested volume figures from the spray exposure tests were used, with substitution of a volume of 0.005 mL (half the Limit of Detection) for individuals where CYA was not detected in urine and 0.025 mL (half the Limit of Quantitation for those with trace amounts of CYA detected in urine. Distribution fitting to the ingested dose data set showed a lognormal distribution produced the best fit (Chi-Sq. 6.6923). The output range was truncated to 0–25 mL based on reported water ingestion estimates from swimming studies. The fitted lognormal distribution had a mean of 0.5560 and standard deviation of 1.576. Monte Carlo simulation was carried out in @Risk Version 5.7.0 using 10,000 iterations to generate a distribution for the adjusted ingested water volume.