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INVITED REVIEW

A systematic review of the use of silicone wristbands for environmental exposure assessment, with a focus on polycyclic aromatic hydrocarbons (PAHs)

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BACKGROUND: Exposure assessment is critical for connecting environmental pollutants to health outcomes and evaluating impacts of interventions or environmental policies. Silicone wristbands (SWBs) show promise for multi-pollutant exposure assessment, including polycyclic aromatic hydrocarbons (PAHs), a ubiquitous class of toxic environmental pollutants.
OBJECTIVE: To review published studies where SWBs were worn on the wrist for human environmental exposure assessments and evaluate the ability of SWBs to capture personal exposures, identify gaps which need to be addressed to implement this tool, and make recommendations for future studies to advance the field of exposure science through utilization of SWBs.
METHODS: We performed a systematic search and a cited reference search in Scopus and extracted key study descriptions.
RESULTS: Thirty-nine unique studies were identified, with analytes including PAHs, pesticides, flame retardants, and tobacco products. SWBs were shipped under ambient conditions without apparent analyte loss, indicating utility for global exposure and health studies. Nineteen articles detected a total of 60 PAHs in at least one SWB. Correlations with other concurrent biological and air measurements indicate the SWB captures exposure to flame retardants, tobacco products, and PAHs.
SIGNIFICANCE: SWBs show promise as a simple-to-deploy tool to estimate environmental and occupational exposures to chemical mixtures, including PAHs.

Keywords: Personal exposure; Polycyclic aromatic hydrocarbons; Volatile organic compounds

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INTRODUCTION

Chemical exposure assessment is a necessary step for risk assessment, and for connecting exposures to health outcomes. Determining exposure is difficult as it varies over space and time, and exposure patterns can depend on personal factors such as age-specific behaviors, especially in children [1]. Exposure assessment at the personal level is preferred and is often assessed through biological samples such as blood or urine, which has the advantage in that biological monitoring evaluates personal exposure by all routes, but is limited for determining individual routes of exposure [2]. In addition, biological samples in field studies may be difficult to obtain and require special handling and shipping of samples [2]. Handwipes, clothing samples, skin adhesive patches, passive and active air samplers have been utilized as less invasive methods for personal exposure assessment. Airborne exposures can be measured using personal air samplers, however, they can be burdensome for study subjects to carry [3]. Silicone wristbands, a pre-cleaned silicone band worn on the wrist, could be a viable tool for exposure assessment because they passively sample a wide range of compounds which can later be extracted and analyzed [4, 5]. These wristbands are also easy to deploy and wear for children and adults and can be worn for periods of days up to weeks [5, 6].

Silicone wristbands were first developed as a tool for personal exposure assessment by O'Connell et al. [5]. They have since been used to measure various compounds including nicotine [7, 8], flame retardants [4, 6, 9–26], pesticides [4–6, 12–14, 19, 22, 24, 27–30], and PAHs [4–6, 13, 14, 17, 26, 31–42]. The potential application of silicone wristband samplers for personal exposure assessments builds on literature applying silicone samplers to evaluate atmospheric and aqueous phase pollutants [28, 32, 39, 43, 44]. Passive air samplers made of silicone have been shown to have higher uptake rates for several indoor volatile organic compounds (VOCs) and semi-VOCs (SVOCs), including organophosphate esters (OPEs), polybrominated diphenyl ethers (PBDEs), polyaromatic hydrocarbons (PAHs), and other compounds compared to established passive air samplers made of polyethylene or styrene divinyl-benzene copolymer [45–47]. Due to the high uptake capacity of silicone to collect air contaminants, a recent field study deployed hanging SWBs inside homes as passive air monitors to measure a variety of SVOCs [48].

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Although there is an increasing body of literature on the use of silicone wristbands to estimate personal exposure, there is no comprehensive review available characterizing the different types of contaminants measured by worn SWBs, the studied populations and regions, and the correlations of SWBs with other personal exposure methods, including biomarkers. A review of this novel scientific literature is required to evaluate the strengths and limitations of SWBs in future environmental health and epidemiological studies. The purpose of this review is to examine and compare studies using SWB when worn on the wrist by human subjects and through this evaluate their current utility for personal exposure assessment. Furthermore, this review evaluates the detection of environmental chemicals by SWBs in comparison to other more accepted exposure measurement techniques. Last, we identify gaps in the current understanding of best practices for field SWB implementation and resulting data interpretation for environmental exposure assessments, and make recommendations for future research needs. To compare across studies and to allow for the potential contribution of sweat and dermal exposure to chemicals absorbed on SWBs, we consider only studies with bracelet-style SWBs worn on the wrist by humans. Excluded studies include those with SWBs hung in the air, other shapes (e.g., silicone military-style 'dog tags' worn by firefighters [49]) and on non-human subjects (e.g., tags for pets to measure OPEs, nBFRs, PCBs, and pesticide levels [22, 50]). We compare study designs and participant types, deployment details such as time worn, chemicals measured and detected, and the relationship of the chemicals detected in the wristband to concurrent measurements such as air sampling or urinary/blood biomarkers of exposure.

An additional focus of this review is on silicone wristbands as a sampler for detecting PAHs, a ubiquitous environmental contaminant. Almost half of the studies using SWBs measured and detected PAHs, which allowed for comparison across studies of various characteristics including detection frequency, time wristband was worn, and chemical uptake. Furthermore, because many PAHs are ubiquitous toxic and carcinogenic compounds, this summary of the literature can help illustrate the benefits of silicone wristbands for understanding widespread personal contact with PAHs, which may ultimately aid in connecting exposure and health outcomes.

PAHs are a class of organic compounds characterized by having two or more benzene rings [51]. They are often byproducts of incomplete combustion and are found throughout the environment in water, soil, and partition between the gas and particle phases in the atmosphere [51]. Some PAHs are carcinogens and/or mutagens; one of the most potent carcinogenic PAHs is benzo(a)pyrene [52]. PAHs are toxic to pregnant women and children, causing DNA adduct formation in mothers and newborns, which is linked to cancer risk [53]. PAH exposure has also been associated with low birth weight [54]. In addition to negative birth outcomes, exposure to PAHs in utero is associated with respiratory and neurocognitive issues [53–56]. The routes of exposure to PAHs are ingestion, inhalation, and dermal [51]. The most frequently measured PAHs are the group of 16 priority PAHs determined by the US EPA [57]. However, the current state of knowledge indicates infrequently measured PAHs, including methyl-, oxy- and nitro-PAHs, should be included in exposure assessments due to their toxicity and abundance [58]. Some of these PAHs are formed during incomplete combustion or oxidation (e.g., chemical, biological, or photo-oxidation) processes [57] and environmental exposure assessments should be more frequent. Therefore, PAHs present a challenging problem for exposure assessment because this chemical class has a multitude of compounds with varying sources and multiple exposure routes.

METHODS

Article selection criteria and search strategy

Included studies had to (1) measure one or more chemical compounds in silicone wristbands (2) present data for silicone wristbands that were worn by human subjects (3) provide primary data, and (4) be published in a peer-reviewed journal. The Scopus database was searched on December 5, 2020 using the following search: (TITLE-ABS-KEY ({silicone wristband} OR {silicone wristbands})) OR (TITLE ABS-KEY(silicone OR PDMS OR polydimethylsiloxane AND wristband)) OR (TITLE-ABSKEY({wrist band} OR wristband AND passive W/2 sampler)) OR (TITLE-ABS-KEY (silicone OR PDMS OR polydimethylsiloxane AND passive W/2 sampler)). Note "passive W/2 sampler" searches "passive" within two words of "sampler." The publication period spanned 1999–2020. We also performed a cited reference search using the first publication that applied silicone wristbands worn by subjects for exposure assessment [5]. The publication period spanned 2014–2020. There was no language limitation placed on the Scopus search.

Document exclusion

We excluded search results if the articles were coded as any of the following: Review, Book, Book Chapter, Conference Paper, Note, Letter, or Erratum. Remaining articles were manually screened by reviewing the title and abstract. Articles that were news, symposium reports, or did not use primary research were excluded along with studies that did not have human subjects wearing silicone wristbands. We also excluded studies that used silicone tubing to capture volatile compounds [45, 59].

The following information was collected from articles passing the selection criteria: the study purpose; study population; sample size; location; analytes; length of time wristbands were worn by subjects; units of measurements reported for wristbands; concurrent air measurements, urine measurements, other environmental measurements (e.g., blood); if PAHs were measured; and if silicone wristbands were shipped at room temperature. Information on PAHs measured and detected was obtained from the article or supporting materials, and from personal communication with the authors [4–6, 13, 14, 17, 31–39]. Additionally, information on international shipping was also obtained via email communication [60–63].

RESULTS

Article selection

The Scopus search terms identified 277 publications, and the document type excluded 23 of those articles. Additional articles were excluded due to a lack of human subjects ($n = 212$) (e.g., PDMS or wristbands used for air measurements only), contained secondary data only ($n = 3$), news article ($n = 1$), or wristbands worn by human subjects but not for exposure detection purposes ($n = 1$). Thirty-nine articles remained at the end of the selection process (Fig. 1).

The cited reference search identified 100 articles that cited O'Connell et al. [5]. Of these, 31 were excluded by document type. The title and abstract of the remaining 69 articles were manually reviewed and exclusions were made due to lack of human subjects ($n = 28$), review article ($n = 5$), news article ($n = 2$), or symposium report ($n = 1$), leaving 33 publications. Of these, all but one were already contained within the final $n = 37$ articles from the Scopus search terms. One additional "in press" article by some of the authors of this review was added. Therefore, the final dataset included 39 unique articles which presented primary data collected from human subjects wearing silicone wristbands [8].

Characteristics of selected studies

Table 1 presents a characterization of the 39 silicone wristband studies measuring a variety of analytes, 19 of which measured PAHs.

Location of study and shipping of samples for analysis

Seven of the studies used wristbands to assess exposure in low- or middle-income countries: Senegal [29], Bangladesh [18],

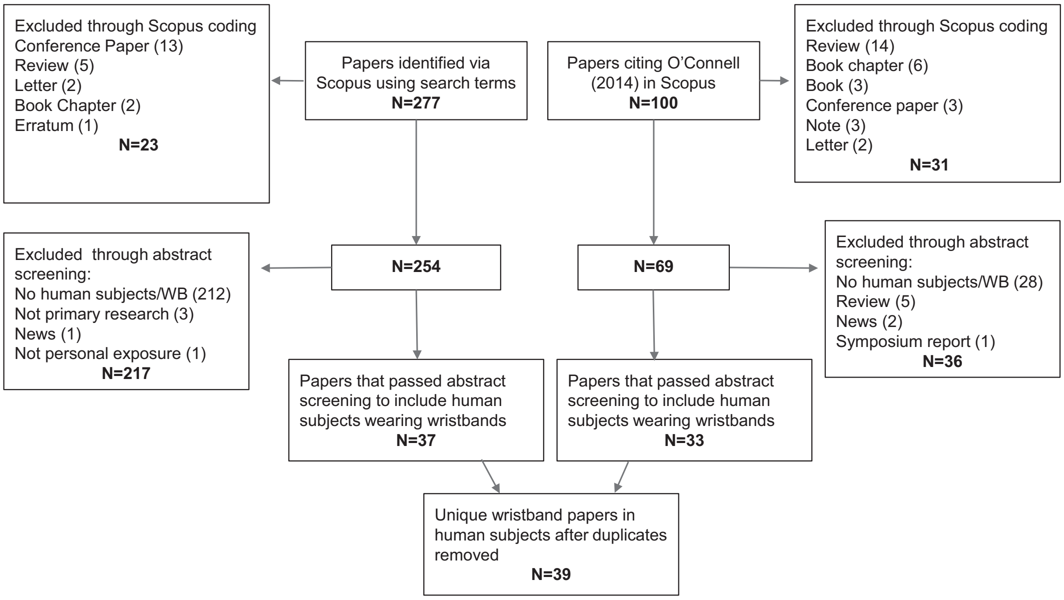

Fig. 1 Study selection process. Process followed for article selection and summary of excluded articles. The center top panels depict the two searches made. The left center box depicts the Scopus search using terms defined in the Methods section and the right center box depicts the cited citation search based on O'Connell, 2014 [5]. The outer panels in the Figure describe the articles excluded for reasons depicted in the boxes, and the bottom box displays the 39 unique articles selected for review. WB: silicone wristband N: number of articles.

Peru [6], South Africa [14], Brazil [13], Chile [38], Dominican Republic [40] (Table 1). Twenty-seven investigations took place in the United States, with five in other high-income countries: Canada [25], Belgium [28], France, Italy [26], Japan [34], and Uruguay [12]. Table 1 provides silicone wristband shipping information for articles that published details or provided details via personal communication. Nine studies shipped or carried wristbands internationally at ambient temperature [6, 12–14, 18, 26, 29, 34, 38]. Six studies shipped wristbands domestically within the United States at ambient temperature [11, 16, 33, 37, 41, 42].

Analytes studied

Silicone wristbands were used to study a variety of analytes across the 39 studies. PAHs were measured in 19 of the articles [4–6, 13, 14, 17, 26, 31–42]. Twenty-four articles measured a single class of analytes [7–11, 15, 16, 18–21, 23, 27–33, 35, 37, 39, 40, 42]; the other 15 measured multiple classes of pollutants [4–6, 12–14, 17, 22, 24–26, 34, 36, 38, 41]. In terms of specific classes of compounds, 13 articles measured pesticides [4–6, 12–14, 22, 24, 27–30, 41] and 20 measured flame retardants [4, 6, 9–24, 26, 36]. More recently, wristbands were utilized to measure nicotine and other smoking-related compounds in two publications [7, 8]. Fourteen of the wristband studies had chemical analyses performed by K. Anderson's laboratory at Oregon State University [4–6, 11, 13, 14, 16, 29–31, 33, 35, 37, 41].

Study participants

Seven studies examined occupational exposure among roofers [5], e-waste dismantlers [18, 25], nail salon technicians [21], firefighters [39, 40], and fishermen [42]. For environmental exposures, children's exposure was the main application for wristbands, as over a third of the studies (12) presented in Table 1 had subjects as young as 3 years old to late teen years [7, 8, 10–12, 14–16, 27, 29, 30, 38]. Additional environmentally exposed subjects in SWB studies included pregnant women [4, 31, 41], Native American Tribal members [37], and susceptible populations such as people with asthma [35]. One area of environmental exposures where silicone wristbands have been applied involves environmental justice communities, such as communities with concerns about nearby oil refineries and gas extraction. Hendryx et al. found that wristbands from the mining group living closer to the site had higher concentrations of total PAHs than the control group [32]. Similarly, Paulik et al. found a negative correlation between PAH concentration in wristbands and distance from an active fracking site [33].

Silicone wristband wearing time

As shown in Table 1, the time periods for which the wristbands have been worn to determine exposure ranged from as little as six hours up to almost five weeks. In a majority of the studies (79%), subjects were asked to wear the wristbands between 1 day to 7 days [4, 7–12, 15–20, 22–24, 26, 28–32, 34–42]. In five articles, wristbands were worn for a week to over a month [5, 6, 14, 27, 33]. In four studies, three of which involved occupational exposure, wristbands were worn for less than 24 h [5, 13, 21, 25].

Units of measurement

In addition to variation in wearing time, units of measurement reported varied across the publications and include nanograms of analyte per wristband (ng/wristband) [4, 5, 7, 17, 23, 31, 34–36], ng per gram of silicone wristband (ng/g SWB), also referred to as ppb [6, 8–10, 12, 13, 15, 19–22, 24, 27–29, 32, 33, 37–42], and in units that incorporate wearing time: ng per gram silicone wristband per day (ng/g-day) [16, 30], ng per hour per wristband (ng/h/wristband) [18], nanogram per hour per square decimeter (ng/h-dm^2) [25]. Other reported units of measurement were the number of chemicals per wristband [14].

Other environmental samples collected

Multiple studies paired silicone wristbands with other measurements including air, handwipe, and biological matrices as well as interviews/surveys. Fourteen publications reported air measurements along with personal wristband data, and of these, four were paired with personal air samplers [4, 8, 25, 31]. In 8 studies non-personal air sampling devices such as active or passive air samplers, low-density polyethylene samplers (LDPE), or nearby EPA monitors [4, 8, 19, 31–35], while others (4 studies) used silicone bands placed in an ambient environment [21, 28, 36, 39]. Three studies measured paired hand wipe samples [10, 23, 36], and 9 took measurements from paired single/multiple spot urine samples or pooled spot urine samples [7, 8, 10, 15, 21–23, 25, 31].

Table 1. Silicone wristband studies and overview of relevant details.

Authors	Purpose	Study pop adult	Study pop Child (yrs)	*n*	Location	SWB measures	Time SWB worn	SWB units	Air	Urine	Other Env	PAHs	SWB ship RT?
Aerts et al. (2018)	Assess pesticide exposure w/ SWBs	CS		30	Belgium	Pesticides	5 D	ng/g S	A, SWBa			N	
Anderson et al. (2017)	Methodological investigation of SWBs	Preg wmn		22	NY, US	FRs, PAHs, PCBs, pesticides, VOCs	2 D	ng/SWB	P, Act			Y	
Baum et al. (2020)	Document firefighters' occupational exposure to PAHs	O, Fire fighters		72	FL, US	PAHs	1 D	ng/g S	A, SWBa			Y	
Bergmann et al. (2017)	Personal exposure to pesticides/other chemicals w/ SWBs	Comm		68	Alto Mayo, Peru	ICs, FRs, PAHs, pesticides	30–34 D	ng/g S				Y	Y, I
Caban-Martinez et al. (2020)	Measure firefighters' exposure to carcinogens during work shift with fire calls	O, Fire fighters		15	Dominican Republic	PAHs	1D	ng/g S				Y	
Craig et al. (2019)	Exposure to SVOCs in nail salon	O, Nail techs		10	MA, US	OPEs, PAs, PHTHs	6–11 H	ng/g S	P, SWBa	Spot		N	
De Vecchi et al. (2019)	Conduct exposome study	Cyclists		2	Brazil	Same as Bergmann	6 H	ng/g S			Hair	Y	Y, I
Dixon et al. (2018)	Compare SWB PAH to air and biomarkers	Preg wmn		22	NY, US	PAHs	2 D	ng/SWB	P, Act	Spot		Y	
Dixon et al. (2019)	Compare multipollutants in SWBs across three continents	Mix	C (3–15)	246	NA, SA, AFR	ICs, FRs, PAHs, PCBs, PCPs, pesticides, PHTHs	Varied	# C/SWB				Y	Y, I
Doherty et al. (2020)	Compare exposure to multipollutants and identify predictors of exposure	Preg wmn		255, 20	NH, US	Same as Dixon (2019)	7 D	ng/g S				Y	Y, D
Donald et al. (2016)	Use SWBs to detect pesticide exposure	Farm fam	C	35	Diender, Senegal	Pesticides	5 D	ng/g S				N	Y, I
Gibson et al. (2019)	OPEs in SWBs compared to urinary metabolites	Mothers	C (3–6)	32 P	NY, US	OPFRs	7 D	ng/g S		Spot		N	
Hammel et al. (2016)	Compare OPFRs in SWBs to other exposure measures	CS		40	NC, US	OPFRs	5 D	ng/SWB		Spot	HW	N	
Hammel et al. (2018)	PBDEs in SWBs compared to serum biomarkers	CS		30	NC, US	BFRs	7 D	ng/g S			Serum	N	
Hammel et al. (2020)	FRs and plasticizers in SWBs compared to other exposure measures		C (3–6)	77	NC, US	NPPZs, OPEs, PHTHs	7 D	ng/g S		SP	Dust, HW	N	
Harley et al. (2019)	Pesticides in SWB in relation to residence near agricultural fields		C LG (14–16)	97	CA, US	Pesticides	7 D	ng/g/day				N	

Table 1 continued

Authors	Purpose	Study pop adult	Study pop Child (yrs)	*n*	Location	SWB measures	Time SWB worn	SWB units	Air	Urine	Other Env	PAHs	SWB ship RT?
Hendryx et al. (2020)	Residential PAH exposure in coal mining communities	Comm		101	WV, KY, VA, US	PAHs	7 D	ng/g S	A, Act			Y	
Kassotis et al. (2020)	Evaluate ability of SWBs to measure mixture of thyroid disrupters	Cancer patients		72	NC, US	BFRs, OPEs, pesticides, PHTHs	7D	ng/g S				N	
Kile et al. (2016)	Demonstrate viability of SWBs for measuring children's exposure to FRs		C (3–5)	72	OR, US	FRs	7 D	ng/SWB/day				N	Y, D
Lipscomb et al. (2017)	FR in SWBs in relation to child's behavior		C (3–5)	72	OR, US	FRs	7 D	ng/g/day				N	Y, D
Manzano et al. (2019)	Spatial and seasonal patterns of SWBs -targeted/ non-targeted analysis		C HS	27	Santiago, Chile	CBPs, CPs, ICs, PAHs, PZs	5 D	ng/g S				Y	Y, I
Nguyen et al. (2020)	Exposure to FRs from recycling e-waste	O, Recycl		45	Quebec, Canada	FRs, OPEs,	6–10 H	ng/h/dm	P, Act, SWBa	Spot	Blood plsma	N	
O'Connell et al. (2014)	First SWBs article - demonstrate use to measure personal exposure	CS; O, Roofers		22, 8	OR, US	CPs, ICs, PAHs, PCPs, pesticides, PHTHs	30 D, 8H, 40 H	ng/SWB	P, SWBa			Y	
Paulik et al. (2018)	PAH exposure from natural gas extraction	Comm		19	OH, US	PAHs	21 D	ng/g S	A, Psv			Y	Y, D
Quintana et al. (2019)	Nicotine in SWB compared to urinary cotinine		C (4–14)	31	CA, US	Nicotine	7D, 2 D	ng/SWB		Spot		N	
Quintana et al. (2020)	Tobacco compounds in SWBs compared to children's exposure		C (4–14)	53	CA, US	Nicotine, TRPs	7D, 2 D	ng/g SWB	P, Psv	Spot		N	
Reche et al. (2020)	Exposure to air pollution during athletic events	Athletes		5	Japan	PAHs, PCPs	106 H	ng/SWB	A, Act			Y	Y, I
Reddam et al. (2020)	Personal exposure to OPEs during commute	Stdnt		88	CA, US	OPEs	5 D	ng/g S				N	
Rohlman et al. (2019a)	Community-engaged environmental Justice study/oil refinery	Tribal		10, 22	WA, US	PAHs	7 D	ng/g S				Y	Y, D
Rohlman et al. (2019b)	Exposure, Location and Lung function (ELF) tool	Asthm		10	OR, US	PAHs	7 D	ng/SWB	A, Act		Spiro-meter	Y	
Romanak (2019)	Method to detect SVOCs in SWBs	CS		10	IN, US	FRs, OPEs, PAHs, PBDEs	7 D	ng/SWB				Y	

Table 1 continued

Authors	Purpose	Study pop adult	Study pop Child (yrs)	*n*	Location	SWB measures	Time SWB worn	SWB units	Air	Urine	Other Env	PAHs	SWB ship RT?
Santiago et al. (2020)	Estimate exposure to PAHs during fishing activities	O, Fishers		9	FL, MS, US	PAHs	1 D	ng/g S				Y	Y, D
Travis et al. (2020)	Measure children's exposure to multiple pollutants in South America		C (6–7.8)	24	Montevideo, Uruguay	FRs, PBDEs, PCBs, pesticides	7 D	ng/g S			Blood	N	Y, I
Vidi et al. (2017)	Link pesticide exposure w/ SWB to genetic damage		C LF (7–9)	10	NC, US	Pesticides	7–14 D	ng/g S			Hair	N	
Wang et al. (2019)	Investigate dermal contribution to SWBs	CS		10	IN, US	nBFRs, OPEs, PAHs, PBDEs	2 D	ng/SWB	P, SWBa		HW	Y	
Wang et al. (2020)	Exposures from recycling e-waste	O, Recycl		15	Dhaka, Bangladesh	FRs, nBFRs, OPEs, PBDEs	1 D	ng/h/SWB			T-shirts	N	Y, I
Wang et al. (2020)	Exposure to FRs w/ SWBs and thyroid function	Comm		101	Appalachia, US	nBFRs, OPFRs, PBDEs	7 D	ng/g S	A, Psv		Blood	N	
Wang et al. (2020)	Compare regional exposures to SVOCs in European countries	Stdnt		40, 31	France, Italy	nBFRs, OPEs, PBDEs, PAHs	5 D	ng/g S				Y	Y, I
Wise et al. (2020)	Compare exposures between dogs and their owners via SWBs and silicone dog tags	CS, Dogs		30	NC, NJ, US	nBFRs, PBDEs, PCBs, pesticides, PHTHs	5 D	ng/g S		SP	SDTs	N	

Asthm volunteers with asthma, *Comm* community members, *CS* convenience sample, *Farm fam* farming families, *Nail techs* nail salon technicians, *O* occupational exposure, *Preg wmn* pregnant women, *Recycl* males who recycle E-waste, *Stdnt* students, *Tribal* tribal members, *C HS* high school students, *C LF 7–9* Latino children 7–9 years old from farmworker households, *C LG 14–16* Latina girls 14–16 years old, *C* included children, *n* the number of subjects wearing wristbands, *P* pairs, *Appalachia, US* 8 communities in rural Central Appalachia, US, *CA, US* California, US, *FL, MS, US* Florida, Mississippi, US, *IN, US* Indiana, US, *Japan* Yokohama 2019 World Relays stadium and warm-up track in Japan, *MA, US* Massachusetts, US, *NA, SA, AFR* North America, South America, Africa, *NC, US* North Carolina, US, *NJ, US* New Jersey, US, *NY, US* New York, US, *OH, US* Ohio, US, *OR, US* Oregon, US, *WA, US* Swinomish Indian Tribal Community in Washington, US, *WV, KY, VA, US* West Virginia, Kentucky, and Virginia, US, *BFRs* brominated flame retardants, *CBPs* combustion products, *CPs* consumer products, *FRs* flame retardants, *ICs* industrial compounds, *nBFRs* novel brominated flame retardants, *NPPZs* non-phthalate plasticizers, *OPEs* organophosphate esters, *OPFRs* organophosphate flame retardants, *PAs* phthalate alternatives, *PBDEs* polybrominated diphenyl ethers, *PCBs* polychlorinated biphenyls (includes dioxins and furans in the case of Dixon (2019)), *PCPs* personal care products, *PHTHs* phthalates, *PZs* plasticizers, *TRPs* tobacco-related products nicotine, cotinine, tobacco-specific nitrosamines, *VOCs* volatile organic chemicals, *D* days, *H* hours, *# C/SWB* number of chemicals per silicone wristband, *ng/g S* nanograms of chemical per gram of silicone wristband, *ng/g/day* nanograms of chemical per gram of silicone wristband per day, *ng/h/SWB* nanograms of chemical per hour per silicone wristband, *ng/h/dm*^{2} nanograms per hour per decimeters^2, *ng/SWB* nanograms of chemical per silicone wristband, *ng/SWB/day* nanograms of chemical per silicone wristband per day, *A* area, *Act* active sampler, *P* personal, *Psv* passive sampler, *SWBa* silicone wristband used for air sampling, *SP* multiple spot samples pooled, *Other env* other environmental samples, *HW* handwipe sample, *SDT* silicone dog tag, *N* no, *Y* yes, *SWB* silicone wristband, *RT* room temperature, *Y, D* yes, mailed domestically, *Y, I* yes, shipped or carried internationally.

Other types of biological samples collected alongside wristbands were blood [12, 19, 25], serum [9], and hair [27]. One article reported on paired silicone wristbands between human subjects and their dogs [22]. Almost all articles had conducted interviews or questionnaires for subjects.

Methodological aspects of silicone wristbands

O'Connell et al. published the first paper demonstrating the use of silicone wristbands as a device to measure personal exposure to various compounds including PAHs [5]. This paper covers details on laboratory method development, as well as deployment in ambient conditions ($n = 22$ people for 30 days) and occupational exposure ($n = 8$ roofers). Various solvent pre-cleaning procedures were compared, with the optimal cleaning method using ethyl acetate, hexane, and methanol. Some more recent articles do not use the chemical cleaning methodology but prepare the wristbands for use through utilization of a vacuum oven [4, 14, 29, 31]. In a recovery experiment, the authors reported that all spiked compounds (PAHs and OPAHs) were extracted and quantitated within 26% of the true value. Extraction of deuterated PAHs infused into wristbands were ≥96% of the spiked amount after two rounds of extraction. Sunlight exposure for a four-hour period did not degrade PAHs on the wristbands, and transport time or temperature variations for up to 72 hours did not lead to a measurable loss of PAHs [5]. In the subjects wearing the wristbands for 30 days at all times including bathing and sleeping, 49 compounds were detected on at least one wristband. Log K_{ow} values of all compounds detected (see Table 1) ranged from −0.07 to 9.49, indicating hydrophilic and hydrophobic compounds can be sampled using silicone wristbands [5].

Anderson et al. describes a methodological study that covered issues of preparation, transport, extraction, and storage [4]. They exposed wristbands to 148 compounds with log octanol-air partitioning coefficients ranging from 2.1 to 13.7 [4]. Fifty of the compounds were PAHs and the remaining 98 were other SVOCs and VOCs. Wristbands were infused with chemicals and stored for 2 days to 6 months at temperatures from −20 °C to +30 °C [4]. The researchers suggested wristbands used for VOC exposure assessment could be stored at up to 30 °C for a maximum of 7 days and were stable up to 3 months at −20 °C. SVOCs were more stable, for at least a month at up to 30 °C, and could be stored at −20 °C over 6 months [4].

PAHs measured and detected using silicone wristbands

A total of 102 PAHs were measured across 19 studies using silicone wristbands (Table 2a–e). Of these, 60 were detected on at least one participant's wristband. Table 2 displays the PAHs grouped by type and indicate which were detected. Molecular weight (MW) of detected PAH compounds ranged from 128.17 to 302.4, log values of the octanol/water partition coefficients (K_{ow}) ranged from 1.71 to 7.71, and log values of the octanol/air partition coefficient (K_{oa}) from 5.01 to 11.76. All 19 studies measured at least four or more of the 16 priority EPA PAHs, and all studies detected multiple PAHs from this list. Additional parent PAHs were measured in 14 of the 19the studies, along with 23 alkylated PAHs. Nitro PAHs, which made up 18 of the 102 total PAHs, were measured in three studies and there were no reported detections [6, 13, 14]. Five studies measured oxy PAHs, however only O'Connell et al. and Doherty et al. reported detections [5, 41].

Table 2a describes the 16 priority EPA PAHs, which were the most commonly measured as a group. Each of the 16 PAHs was detected in at least one study, with phenanthrene [17, 33–37, 39, 40, 42] measured in all studies and detected in 18 of the 19 studies which measured this compound. Acenaphthylene, fluorene, pyrene and fluoranthene were measured in 18 of the 19 studies, and these four compounds were detected in 17 of the 18 studies which measured them (Table 2a) [13, 17, 33–37, 39, 40, 42]. Benzo[a]pyrene, one of the most toxic PAHs, was measured across 15 articles and detected in 9 of these 15 studies [4, 5, 31, 33, 35, 37, 39–41]. Three of the studies detected all 16 PAHs in at least one wristband [4, 31, 40]. Paulik et al. reported detecting 15 of the 16 PAHs and of all studies, they had the most PAHs detected across all wristbands (20 PAHs), likely due to this being a study of communities near gas extraction sites [33].

Twenty-five additional parent PAHs and one hetero-PAH (sulfur-containing PAH, dibenzothiophene) were measured by one or more of the studies and are listed in Table 2b. Over 60% (16) of these PAHs were detected in wristbands from at least one participant. Out of the 19 studies that measured PAHs, 10 of them measured dibenzothiophene and triphenylene. Dibenzothiophene was the most frequently detected, as it was detected in 9 of the 10 studies that measured this compound. Two of the studies reported detecting dibenzothiophene in 100% of wristband samples (sample sizes 19 and 11) [33, 35]. Triphenylene was the next most frequently detected, in 8 of 10 publications that measured this compound.

Alkylated PAHs, less commonly measured in environmental studies, were measured in 15 of the 19 articles (Table 2c). O'Connell et al. measured 23 alkylated PAHs and had over 40% detection rate [5]. Multiple studies reported 100% detection rate of several alkylated PAHs across all wristband samples: 2-methylnaphthalene, 1-methylnaphthalene, 2-ethylnaphthalene, 2,6-dimethylnaphthalene, 1,6-dimethylnaphthalene, 1,4-dimethylnaphthalene, 2-methylphenanthrene, 2-methylanthracene, 3,6-dimethylphenanthrene, 1-methylpyrene, and retene [33, 35, 37, 40, 42]. The most commonly detected alkylated PAHs across all studies were 1-methylnaphthalene and 2-methylnaphthalene.

Table 2d and 2e display nitro and oxy PAHs, respectively, which are of interest due to their toxicity. Nitro PAHs were not commonly measured; only 3 studies attempted measurements, and there were no detections [6, 13, 14]. Oxy PAHs were measured across the same three studies as nitro PAHs and in two additional studies [5, 41]; of these, only O'Connell et al. and Doherty et al. reported detections of oxy PAHs. Both reported detecting 9-fluorenone, O'Connell et al. also reported benzofluorenone (not measured by Doherty et al.), and Doherty et al. also reported detections of benzo[b]fluorene, 1,4-naphthoquinone, 4H-cyclopenta[def]phenanthren-4-one and benzo[a]pyrene-7,8-dione [5, 41].

A summary compiling the list of all PAHs measured and detected in SWBs across the 19 studies is presented in Supplementary Table S1. Supplementary Fig. S1 graphically presents the maximum time wristbands were worn along with the maximum percentage of PAHs detected for all the studies that measured PAHs, with the exception of O'Connell et al. [5] and Dixon et al. [14] since the time worn varied. Supplementary Fig. S1 also specifies whether wristbands in each study were worn in occupational or general environmental conditions. No relationship was observed between time worn and detection frequency, however, the exposure levels and experimental design differed among studies.

Associations between analytes in SWBs and personal exposure measurements

One way to evaluate the ability of SWBs to function as a personal exposure assessment is to examine correlations of SWB-based measures with those from other better established methods. In Supplementary Table S2, we present published correlations between wristband results and those of other personal measurements from the same individual. Most of the reviewed studies ($n = 8$) that evaluated SWBs in comparison to other personal samplers targeted flame retardants such as OPFRs/OPEs [10, 22, 23, 25, 36, 64], PBDEs, and nHFRs/nBFRs [18, 25, 36]. Personal samples include those external to the subject such as personal air samplers (either active or passive), and handwipes, and those reflecting internal dose such as biomarkers in urine or blood.

Table 2. Polycyclic aromatic hydrocarbons (PAHs) measured and detected in silicone wristbands a. 16 EPA PAHs$^+$. b. Additional Parent PAHs. c. Alkylated PAHs. d. Nitro PAHs. e. Oxy PAHs.

PAH	CAS#	MW (g/mol)	Kow[a]	Koa[b]	Study number																		
					1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
a. 16 EPA PAHs$^+$																							
Naphthalene	91–20–3	128.17	3.30	5.17	X	X	X	Ӿ	x	X	X	X		X	X	Ӿ		Ӿ	Ӿ		Ӿ		
Acenaphthylene	208–96–8	152.19	3.94	6.56	X	X	X	Ӿ	x	X	X	X		X	X	X	Ӿ	X	X	Ӿ	Ӿ	Ӿ	X
Acenaphthene	83–32–9	154.21	3.92	6.33	X	X	x	X	x	X	x	X		X	X	X	Ӿ	X	X	X	X	X	x
Fluorene	86–73–7	166.22	4.18	6.84	X	X	X	Ӿ	x	X	X	X	X	X	X	Ӿ	Ӿ	X	Ӿ	Ӿ	X	Ӿ	X
Anthracene	120–12–7	178.23	4.47	7.55	X	X	X	X	x	X	X	X	X	X	X	Ӿ		X	x	X	X	Ӿ	X
Phenanthrene	85–01–8	178.23	4.45	7.55	X	X	X	Ӿ	x	X	X	X	X	X	X	Ӿ	Ӿ	Ӿ	Ӿ	Ӿ	Ӿ	Ӿ	Ӿ
Fluoranthene	206–44–0	202.25	5.16	8.86	X	X	X	Ӿ	x	X	X	X	X	X	X	Ӿ		X	X	Ӿ	X	Ӿ	X
Pyrene	129–00–0	202.25	4.88	8.86	X	X	X	Ӿ	x	X	X	X	X	X	X	Ӿ		X	X	Ӿ	Ӿ	Ӿ	Ӿ
Benz[a]anthracene	56–55–3	228.3	5.60	9.37	X	X	X	Ӿ	x	X	X	X	X	X	X	X		X	X	Ӿ	X	X	X
Chrysene	218–01–9	228.3	5.81	9.37	X	X	X	Ӿ	x	X	X	X	X	X	X	Ӿ		X	X	X	X	X	X
Benzo[a]pyrene	50–32–8	252.3	5.78	8.64	X	X	x	X	x	X	x	X		x	X	X		X	X		x		x
Benzo[k]fluoranthene	207–08–9	252.3	6.11	9.38	X	X	x	X	x	X	x	X	X	x	x	X		X	x	x	x	Ӿ	x
Benzo[b]fluoranthene	205–99–2	252.3	6.13	9.61	X	X	x	Ӿ	x	X	x		X	x	X	Ӿ		X	x	x			x
Indeno[1,2,3-c,d]pyrene	193–39–5	276.3	6.76	11.72	X		x	X	x	X	x			x	x	X		X	x		X		X
Benzo[ghi]perylene	191–24–2	276.3	6.63	11.72	X	X	x	X	x	X	x			x	X	X		X	X		x		x
Dibenz[a,h]anthracene	53–70–3	302.4	6.72	11.69	X		x	X	x	X	x	X		x	x	x		x	x		X		x
Total number of PAHs measured					16	14	16	16	16	16	16	13	9	16	16	15	4	16	16	11	15	10	15
Total number of PAHs detected in 1 wristband					16	14	9	16	0	16	9	13	9	10	13	14	4	15	11	9	12	10	9
Total number of PAHs detected in all wristbands					–	–	–	8	–	–	–	–	–	–	–	8	4	2	3	6	4	7	2
b. Additional Parent PAHs																							
Dicyclopentadiene	77–73–6	132.2	3.71	4.44							x												
Dibenzothiophene[s]	132–65–0	184.26	4.44	7.50	X	X	X		x	X	X				X	Ӿ		X	Ӿ				
Benzo[a]fluorene	238–84–6	216.28	5.54	7.88	X	X	X		x	X	X	X			x	X		X	x				
Benzo[b]fluorene	243–17–4	216.28	5.77	8.52	X	X	X		x	X	X	X			x	X		X	x				
Benzo[c]fluorene	205–12–9	216.28	5.38	8.52	X	X	x		x	X	x				X	X		X	X				
Cyclopenta[c,d]pyrene	27208–37–3	226.3	5.70	9.63	X	X	X		x	X	X				x	X		X	x				
Triphenylene	217–59–4	228.3	5.49	9.37	X	X	X		x	X	X				x	X		X	X				
Benz[j]&[e]aceanthrylene	202–33–5 199–54–2	252.3	5.70 6.00	6.35 10.90			x		x	x	x				x	x		x	x				
Benzo[e]pyrene	192–97–2	252.3	6.43	10.34	X	X	X		x	X	X				X	X		X	x	x			x
Benzo[j]fluoranthene	205–82–3	252.3	6.30	10.34	X	X	x		x	X	x				x	X		X	X				
Perylene	198–55–0	252.3	6.05	10.34				x							x								
Anthanthrene	191–26–4	276.3	7.04	11.72	x		x		x	x	x				x	x		x	x				
Benzo[a]chrysene	213–46–7	278.3	7.10	11.69	X		x		x	X	x				x	x		X	x				
Coronene	191–07–1	300.4	7.64	11.69	X		x		x	X	x				x	X		X	X				x
Dibenzo[a,h]pyrene	189–64–0	302.4	7.39	11.69			x		x	x	x				x	x		x	x				
Naphtho[1,2-b]fluoranthene	111189–32–3	302.4	7.47	11.69	X		x		x	X	x				x	x		X	x				
Naphtho[2,3-j]fluoranthene	205–83–4	302.4	7.37	11.69	X		x		x	X	x				x	x		X	x				
Dibenzo[a,e]fluoranthene	5385–75–1	302.4	7.49	11.69	X		x		x	X	x				x	x		x	x				

Table 2 continued

PAH	CAS#	MW (g/mol)	Kow[a]	Koa[b]	Study number																		
					1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
Dibenzo[a,l]pyrene	191–30–0	302.4	7.71	11.69	X		x		x	X	x				x	x		x	x				
Naphtho[2,3-k]fluoranthene	207–18–1	302.4	5.11	10.26			x		x	x	x				x	x		x	x				
Naphtho[2,3-e]pyrene	193–09–9	302.4	7.38	11.69			x		x	x	x				x	x		x	x				
Dibenzo[a,e]pyrene	192–65–4	302.4	6.96	11.76	X		x		x	X	x				x	x		x	x				
Dibenzo[e,l]pyrene	192–51–8	302.4	7.52	11.69	X		x		x	X	x				x	x		X	x				
Naphtho[2,3-a]pyrene	196–42–9	302.4	7.34	11.69			x		x	x	x				x	x		x	x				
Benzo[b]perylene	197–70–6	302.4	7.63	11.69			x		x	x	x				x	x		x	x				
Dibenzo[a,i]pyrene	189–55–9	302.4	7.39	11.69	X		x		x	X	x				x	x		x	x				
Total number of PAHs measured					18	8	24	1	24	24	25	2	0	0	25	24	0	24	24	1	0	0	2
Total number of PAHs detected in 1 wristband					17	8	6	0	0	17	6	2	0	0	3	9	0	13	5	0	0	0	0
Total number of PAHs detected in all wristbands					–	–	–	–	–	–	–	–	–	–	–	1	–	–	1	–	–	–	–
c. Alkylated PAHs																							
2-Methylnaphthalene	91–57–6	142.2	3.86	5.83	X	X	X	~~X~~	x	X	X	X		X	X	~~X~~		~~X~~	~~X~~		~~X~~		
1-Methylnaphthalene	90–12–0	142.2	3.87	5.01	X	X	X	~~X~~	X	X	X	X			X	~~X~~		~~X~~	~~X~~		~~X~~		
2-Ethylnaphthalene	939–27–5	156.22	4.38	6.07	X	X	X		x	X	X				x	~~X~~		X	~~X~~				
2,6-Dimethylnaphthalene	581–42–0	156.22	4.31	5.96	X	X	X		X	X	X	X			x	~~X~~		X	~~X~~				
1,6-Dimethylnaphthalene	575–43–9	156.22	3.92	5.79	X	X	X		X	X	X	X			X	~~X~~		X	~~X~~				
1,4-Dimethylnaphthalene	571–58–4	156.22	4.37	5.79	X	X	x		x	X	x	X			x	~~X~~		X	X				
1,5-Dimethylnaphthalene	571–61–9	156.22	4.38	5.96	X	X	x		x	X	x				x	X		X	X				
1,2-Dimethylnaphthalene	573–98–8	156.22	4.31	5.96	X	X	X		x	X	X	X			X	X		X	X				
1,8-Dimethylnaphthalene	569–41–5	156.22	4.26	5.96	x	x	x		x	x	x	X			x	x		x	x				
1,3-Dimethylnaphthalene	575–41–7	156.22	4.42	5.96											x								
2,6-Diethylnaphthalene	59919–41–4	184.28	4.75	7.25	X	x	X		x	X	X	X			x	X		X	X				
2-Methylphenanthrene	2531–84–2	192.25	4.87	8.41	X	X	X		x	X	X	X			X	~~X~~		~~X~~	X				
2-Methylanthracene	0613–12–7	192.25	5.00	8.41	X	X	X		x	X	X	X			X	~~X~~		X	x				
1-Methylphenanthrene	832–69–9	192.25	5.08	7.55	X	X	X		x	X	X	X			X	X		X	X				
9-Methylanthracene	779–02–2	192.25	5.07	7.55	x		X		x	x	X				x	x		x	x				
3,6-Dimethylphenanthrene	1576–67–6	206.28	5.20	7.87	X	X	X		x	X	X	X			X	~~X~~		X	X				
2,3-Dimethylanthracene	613–06–9	206.28	5.19	7.87	X		x		x	X	x				x	X		x	x				
9,10-Dimethylanthracene	781–43–1	206.28	5.69	7.87	x		x		x	x	x				x	x		x	x				
1-Methylpyrene	2381–21–7	216.28	5.47	9.00	X	X	X		x	X	X	X			X	~~X~~		X	X				
Retene	483–65–8	234.3	5.54	8.53	X	X	X		x	X	X	X			X	~~X~~		X	X	X			x
6-Methylchrysene	1705–85–7	242.3	5.72	9.82	X	X	x		x	X	x	X			x	X		X	x				
5-Methylchrysene	3697–24–3	242.3	5.72	9.82		X	x		x	x	x				x	x		x	x				
7,12-Dimethylbenz[a]anthracene	57–97–6	256.3	5.80	10.25	X	X	x		x	X	x				x	X		x	X				
Total number of PAHs measured					21	19	22	2	22	22	22	15	0	1	23	22	0	22	22	1	2	0	1
Total number of PAHs detected in 1 wristband					18	17	14	2	3	18	14	15	0	1	10	18	0	16	15	1	2	0	0
Total number of PAHs detected in all wristbands					–	–	–	2	–	–	–	–	–	–	–	11	–	3	5	–	2	–	–

Table 2 continued

PAH	CAS#	MW (g/mol)	Kow[a]	Koa[b]	Study number																		
					1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
d. Nitro PAHs																							
1-Nitronaphthalene	86–57–7	173.17	3.17	5.77			x		x		x												
2-Nitronaphthalene	581–89–5	173.17	3.24	6.08			x		x		x												
5-Nitroacenaphthene	602–87–9	199.2	3.85	7.46			x		x		x												
2-Nitrofluorene	607–57–8	211.22	3.38	7.55			x		x		x												
2-Nitroanthracene	3586–69–4	223.23	4.04	8.02			x		x		x												
3-Nitrophenanthrene	17024–19–0	223.23	4.04	8.02			x		x		x												
9-Nitroanthracene	602–60–8	223.23	4.78	7.37			x		x		x												
9-Nitrophenanthrene	954–46–1	223.23	4.02	8.02			x		x		x												
1-Nitropyrene	5522–43–0	247.25	5.06	9.32			x		x		x												
2-Nitropyrene	789–07–1	247.25	4.70	9.32			x		x		x												
3-Nitrofluoranthene	892–21–7	247.25	3.98	4.61			x		x		x												
6-Nitrochrysene	7496–02–8	273.3	5.17	10.57			x		x		x												
7-Nitrobenz[a]anthracene	20268–51–3	273.3	4.01	9.40			x		x		x												
3-Nitrobenzanthrone	17117–34–9	275.26	4.24	9.88			x		x		x												
1,3-Dinitropyrene	75321–20–9	292.24	4.12	10.59			x		x		x												
1,6-Dinitropyrene	42397–64–8	292.24	4.25	10.61			x		x		x												
1,8-Dinitropyrene	42397–65–9	292.24	4.27	10.61			x		x		x												
6-Nitrobenzo[a]pyrene	63041–90–7	297.3	5.81	11.70			x		x		x												
Total number of PAHs measured					0	0	18	0	18	0	18	0	0	0	0	0	0	0	0	0	0	0	0
Total number of PAHs detected in 1 wristband					0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Total number of PAHs detected in all wristbands					–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
e. Oxy PAHs																							
1-Hydroxynaphthalene	90–15–3	144.17	2.85	8.36			x		x		x	X											
1,4-Naphthoquinone	130–15–4	158.15	1.71	5.72							x	X											
9-Fluorenone	486–25–9	180.2	3.58	7.51			x		x		x	X			X								
Acenaphthenequinone	82–86–0	182.17	1.95	7.38			x		x		x												
4H-Cyclopenta[def]phenanthren-4-one	5737–13–3	204.22	3.87	8.96			x		x		x	X											
1,4-Anthraquinone	635–12–1	208.21	3.53	8.51			x		x		x												
9,10-Anthraquinone	84–65–1	208.21	3.39	8.41			x		x		x												
9,10-Phenanthrenequinone	84–11–7	208.21	2.52	8.51			x		x		x												
Phenanthrene-1,4-dione	569–15–3	208.21	3.19	8.41			x		x		x												
2-Methyl-9,10-anthraquinone	84–54–8	222.24	3.34	8.94							x												
Benzofluorenone	76723–60–9	230.26	4.73	10.30											X								
Benzanthrone	82–05–3	230.26	4.81	9.69			x		x		x												
Benzo[a]fluoren-11-one	479–79–8	230.26	4.75	9.69			x		x		x												
Naphthanthrone	82–05–3	230.26	4.81	9.69			x		x		x												
5,12-Naphthacene-quinone	1090–13–7	258.3	4.29	9.74			x		x		x												
Benz[a]anthracene-7,12-dione	2498–66–0	258.3	4.40	9.74			x		x		x												

Table 2 continued

PAH	CAS#	MW (g/mol)	Kow[a]	Koa[b]	Study number																		
					1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
Benzo[c]phenanthrene[1,4]quinone	109699–80–1	258.3	3.96	9.82			x		x		x												
1,6-Benzo[a]pyrene-quinone	3067–13–8	282.3	4.31	11.57			x		x		x												
Benzo[a]pyrene-7,8-dione	65199–11–3	282.3	4.60	11.72			x		x		x	X											
Total number of PAHs measured					0	0	18	0	16	0	18	5	0	0	2	0	0	0	0	0	0	0	0
Total number of PAHs detected in 1 wristband					0	0	0	0	0	0	0	5	0	0	2	0	0	0	0	0	0	0	0
Total number of PAHs detected in all wristbands					–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–

[+]EPA. Appendix A to 40 CFR, Part 423–126 Priority Pollutants.

x measured, *X* detected in one or more wristband, ~~X~~ detected across all wristbands in study, [1]Anderson et al. (2017); [2]Baum et al. (2020); [3]Bergmann et al. (2017); [4]Caban-Martinez et al. (2020); [5]De Vecchi et al. (2019); [6]Dixon et al. (2018); [7]Dixon et al. (2019); [8]Doherty et al. (2020); [9]Hendryx et al. (2020); [10]Manzano et al. (2019); [11]O'Connell et al. (2014); [12]Paulik et al. (2018); [13]Reche et al. (2020); [14]Rohlman et al. (2019A); [15]Rohlman et al. (2019B); [16]Romanak et al. (2019); [17]Santiago et al. (2020); [18]Wang et al. (2019), [19]Wang et al. (2020), *CAS#* Chemical Abstracts Service Registry Number, *MW* molecular weight, *s* sulfur-containing hetero-PAH.

[a]Octanol-water partition coefficient (log).

[b]Octanol-air partition coefficient (log).

Associations with biomarkers

Dixon et al. [31] found that PAH urinary metabolites were correlated with the PAH levels found in SWBs (*rho* = 0.47–0.66). In addition, Quintana et al. reported high correlations between SWB wristband levels of nicotine and cotinine (worn for 2 or 7 days) and urinary cotinine, a metabolite of nicotine ($R^2 =$ 0.74–0.80, *rho* = 0.84–0.91) [7, 8]. Several OPFRs and OPEs detected in SWBs were significantly positively associated with urinary metabolites (Supplementary Table S2) [22, 23, 36, 64].

Associations with personal air samplers and handwipes

There were significant positive associations (*rho* = 0.49–0.71; $R^2 =$ 0.17–0.41) between PAH levels detected using personal air samplers and SWBs levels [4, 31, 36]. Additionally, Quintana et al. found that personal air nicotine concentrations as measured by the Hammond passive air sampler measured over 7 days were significantly correlated with SWB nicotine (*rho* = 0.71, $p < 0.001$) [8]. Regarding SWBs and handwipes, Wang et al. [36] found significant associations among PAHs ($R^2 = 0.58$), OPEs ($R^2 = 0.40$), PBDEs ($R^2 = 0.73$), and nBFRs ($R^2 = 0.67$) as measured by handwipes vs. SWBs, Wang et al. also found significant associations between concentrations detected in SWBs and brooches worn within the breathing zone for PAHs ($R^2 = 0.47$), OPEs ($R^2 =$ 0.73), PBDEs ($R^2 = 0.58$), and nBFRs ($R^2 = 0.51$) (Supplementary Table S2). However, no significant associations were found between air concentrations of PAHs ($R^2 = 0.07$) or PBDEs ($R^2 =$ 0.006), as measured by the standard OVS sampler and SWB, although significant associations were found for OPEs ($R^2 = 0.63$) and nBFRs ($R^2 = 0.76$).

Associations with area samples

Two other studies compared PAHs in worn SWB with air measurements of PAHs area samples. PAHs detected in outdoor air with active PUF samplers had significant but modest correlations (*rho* = 0.22–0.24, $p < 0.005$) with SWBs worn by participants [32]. PAHs collected by passive low-density polyethylene (LDPE) air samplers in an area near to a natural gas extraction site were significantly correlated with the ∑PAH levels found in SWBs (*rho* = 0.64, $p < 0.005$) [33].

DISCUSSION

Silicone wristbands as a measure of personal exposure

Silicone wristbands sorbed a variety of compounds, including PAHs, pesticides, SVOCs, and those in consumer products. The limited numbers of studies that compared levels of chemicals in SWB with personal exposure measures of the same chemical reported moderate to high significant positive correlations, even with small sample sizes [4, 10, 14, 18, 22, 23, 25, 36, 64]. Quintana et al. reported high correlations (*rho* = 0.9) between nicotine in a child's worn wristband and their urinary cotinine concentration, the metabolite of nicotine and the gold standard for tobacco product exposure assessment, as well as a significant difference between groups of children with differing exposure patterns (secondhand smoke and/or e-cigarette vapor) [8]. This indicates that SWBs appear to measure personal exposure, at least for tobacco analytes. However, a gap in our knowledge remains as to whether the silicone wristband is measuring exposures external to the individual (such as personal air concentrations) or presents a non-intrusive measure of internal body burden, or is a combination of both that may depend on exposure types, routes, and levels. (See *Gaps and recommendations*, below).

The detection of PAHs in wristbands across multiple studies are evidence of the ability of silicone wristbands to sorb these compounds, with many studies detecting specific PAHs in every wristband sample [17, 33–37, 40, 42]. Both high molecular weight PAHs that are typically found in the atmospheric particle phase and low molecular weight PAHs predominantly in the gas phase were detected on the SWBs. In all, 60 PAHs were detected across all studies reviewed. Increased measurement of PAHs in addition to the 16 EPA PAHs in personal wristband samples would increase knowledge of the sources and levels of additional toxic PAHs in environmental exposures [57].

SWB applications

Ease of deployment and shipping are important considerations when extending the benefits of exposure assessment to populations and communities with barriers to conventional exposure assessment. A feature that can make these wristbands samplers especially useful is that they appear to be able to be shipped by mail or transported by airplane at ambient temperatures without significant loss of analytes, both domestically [11, 16, 33, 37, 41, 42] and internationally [6, 12–14, 18, 26, 29, 34, 38, 60–63]. They are also easy to deploy without extensive training. The SWBs have been deployed to assess exposure of children [7, 8, 10–12, 14–16, 27, 29, 30, 38], which substantiates the acceptability of this exposure tool in this group. Silicone wristbands have also been used by tribal communities [37] and have been used to support environmental justice investigations [32, 33]. Occupational applications of wristbands have produced findings to support exposure reduction activities [39].

Gaps and recommendations

This review identified areas of future research to address current gaps in knowledge regarding the SWB.

Uptake of chemicals into the worn silicone wristband through air. One gap is characterizing the source of the chemicals and therefore the route(s) of exposure measured by the wristband for the chemicals of interest. It has been documented that silicone wristbands can capture exposures from air [4, 5, 45]. The silicone wristband has been used for air sampling [48, 65] and silicone-based air sampling devices are an area of active research [48, 65, 66]. The theory and factors affecting uptake of chemicals from airborne exposures into silicone sampling devices are reviewed in Wania et al. and Nguyen et al. [25, 65]. One consideration for passive samplers is whether they are sampling during a kinetic uptake phase or when chemicals are in equilibrium with the sampler. Studies of silicone (PDMS) in sheets hung in the air as air samplers indicate that PDMS has a high sorptive capacity [67] and the silicone wristband sampler could be sampling mainly in the kinetic phase for many chemicals. Partition coefficients for PDMS:air were estimated for a wide range of chemicals [67]. For all but a few chemicals, based on the estimated partition coefficients and assuming the silicone sheet sampler had a volume of 10 cm^3 and a sampling rate of 1.7 m^3/day (Okeme et al. [47, 67]), the authors report the time to 25% equilibrium capacity was between 1 day for alpha-hexachlorocyclohexane (a-HCH) to around 500 years for tris (4-tert-butylphenyl) phosphate (TTBPP). For PAHs listed in the publication, the estimated time to 25% equilibrium capacity ranged from 3 days for phenanthrene and anthracene (log K PDMS:air of ~6) to 990 days for the higher molecular weight benzo[a]pyrene (log K PDMS:air estimated at 8.7). Silicone wristbands range in size and thickness, but a 'large' wristband (20.3 cm long × 1.3 cm wide × 0.2 cm thick) has a surface area of 26.4 cm^2 and a volume of 5.3 cm^3. Therefore, although the sampling flow rate is not known for SWBs, and likely will vary, if we apply the analysis and flow rate of Okeme et al., sorptive capacity will likely be about half that of the silicone sheet example in Okeme et al. [67] (~1.5 to 495 days to 25% of equilibrium capacity). The preparation of the PDMS might also affect sorptive capacity and the PDMS:air partition coefficients [68]. For PAHs specifically, an overall generic gas-phase uptake of PAHs on commercial SWBs hung in a chamber was estimated to be $7.6 \pm 1.3\ m^3 d^{-1}\ dm^{-2}$ at an air velocity of 1.3 $cm\ s^{-1}$. However, the uptake rate is still undetermined for SWBs worn on the wrist by individuals as personal passive exposure tools [45]. Tromp et al. demonstrated that the SWBs uptake rate depends heavily on the air velocity, which is usually unknown and not considered in studies where participants wore the SWBs, but is likely higher when worn on the wrist than when air samplers are hung indoors [45]. Therefore, additional controlled studies are needed relating air uptake rates of the SWB in a chamber with known concentrations of the chemical at known flow rates to provide theoretical sampling rates under controlled conditions, and these will help inform sampling study design. However, these studies will not provide information on actual sampling rates when worn by participants given variations in air flow, variations in the concentration of the chemicals across the varied microenvironments the participant encounters throughout the day, and variation in chemicals on the surface of the wristband (e.g., lipids from the body). For higher molecular weight PAHs and other SVOCs, even those mostly bound to particles can partition into air in the indoor environment [69, 70]. Therefore, it may be difficult to parse out whether direct particle partitioning into wristband or partitioning from air to wristband is the source of certain compounds.

Standardization of silicone wristband reporting. A standardized approach to using the wristband and reporting these variables and units of measurement is necessary as differences in sampling duration and units used to report chemical concentrations make it difficult to compare results across studies. In current studies, the type of exposure, sampling duration, and the units used to report PAHs concentration vary across studies, making it difficult to determine sampling parameters [4, 31, 32, 36, 39]. Given the wide variation in units reported for the wristbands (ng/SWB, ng/g SWB, and other units incorporating time), comparisons across studies and understanding of uptake factors would be facilitated by reporting data in multiple units, including ng/surface area of the wristband. We recommend that all studies report data in ng/g wristband as well as ng/wristband surface area and report average weights, surface areas and volumes of the wristbands before deployment. Only Nguyen et al. reported silicone wristband results in units related to the surface area of the wristband, even though the theory of sampling and the sampling rates reported for airborne silicone samplers are in units related to surface area [25].

Uptake of chemicals into the wristband by routes other than uptake from air. In addition to uptake from air, and therefore the potential to measure exposures through inhalation, other potential routes of exposure potentially captured by the SWB that have been suggested in the literature so far are dermal, ingestion, and exposure by all routes (sweat) [28, 36]. Aerts et al. detected almost twice as many types of pesticide residues on personal wristbands worn by individuals as on the paired wristband that was exposed only to air at the home of the person wearing the wristband [28]. They argued that the personal wristbands likely detect exposure routes other than air; possibly ingestion through water or diet or dermal exposures where compounds are absorbed from the skin or sweat into the wristband [28]. Wang et al. found that correlations between wristbands and hand wipes were generally higher than between air samples and wristbands, suggesting that dermal exposure contributes to the exposure captured by wristbands in addition to air exposures [36]. They indicate that wristbands may be a good tool for measuring integrated inhalation and dermal exposure of PAHs [36]. Quintana et al. have also suggested that wristbands may absorb compounds excreted through sweat, as the absorbed dose of nicotine (urinary cotinine) was very highly correlated with nicotine and cotinine in the wristbands, and nicotine and cotinine are excreted in sweat [7, 8]. Manzano et al. applied non-targeted analysis to wristbands collected in students and detected dietary products such as caffeine in the SWB [38]. It is possible that routine measurement of a compound such as a common dietary component could be used in the future as a way to verify wearing of the wristband in personal exposure studies. We recommend that if possible, studies could combine a wristband worn on the wrist with a silicone wristband worn outside of clothing, hung in the home, or the silicone brooch [47] to investigate the contribution of air concentrations to uptake by the silicone wristband. The use of a silicone brooch within the breathing zone can help determine the concentration of contaminants to which participants may be exposed via inhalation. In addition, a handwipe sample or sweat sample would contribute to our knowledge of the potential contribution of skin concentrations to chemicals detected in the SWB. Additional research is needed on the contribution of dermal exposures, sweat, and/or handling objects such as food to measured compounds in the wristband. Further investigation into the partitioning of chemicals by the above routes of exposures into the silicone wristband is needed and would improve interpretation of data collected with these devices and focus their use in research studies.

Wearing time for the SWB worn on the wrist. Another gap is in regard to the length of time silicone wristbands should be worn. As noted in the section above on *Uptake of chemicals into the worn silicone wristband through air*, a duration of a few days to a week should be suitable for capturing chemicals through the inhalation route of exposure, and SWBs will likely be operating as a kinetic sampler for most chemicals (not reaching equilibrium concentrations). Of course, the time worn would typically be shorter for environments with higher concentrations, such as occupational settings, and would be longer for environments with lower concentrations. Even the short wearing times of 1 to 2 days had relatively high detection frequencies for PAHs (Supplementary Fig. S1), and no trend of increasing detection frequency with increased wearing time was evident. However, the short wearing times of <2 days were mainly occupational studies, which may not reflect the ability of the SWB to detect potentially lower environmental exposures during a short wearing time. The only non-occupational study where SWBs were worn less than 1 day was a study of only $n = 2$ cyclists where in a 6 h wearing time, only three PAHs were detected (3.1%) [13] (Supplementary Fig. S1); this illustrates that more non-occupational studies with shorter wearing times are needed to assess minimum wearing times. Anderson et al. concluded that a few lower molecular weight PAH compounds reached equilibrium in the wristband when worn for less than 7 days by pregnant women, as 1 day and 7 day levels were similar (although the data is not presented in the article) [4]. However, as detailed above, this may not be the case for many compounds [67]. Wang et al. suggested that wristbands could measure particle-phase compounds, in addition to vapor phase compounds, as reported by Okeme et al. [36, 71]. Variability in exposure levels over time should be also considered when selecting the final wearing time. For example, if exposures are intermittent and vary over a week, then wearing the SWB for a week rather than 2 days would help to capture the range of exposures and smooth out variability. If possible, multiple wristbands could be given to a subject, and single wristbands removed and stored at regular intervals, to help determine the question of exposure variability and optimal sampling length.

Shipping and storing silicone wristbands. Shipping and storing silicone wristbands at temperatures as high as 30 °C for a week for VOCs and up to a month for many SVOCs seems to be acceptable [6, 11–14, 16, 18, 26, 29, 33, 34, 37, 38, 41, 42, 60–63]. One approach to determine acceptable conditions for shipping and storage modality regarding the specific compounds being studied, if no data exists, is to conduct a pilot study in which a subset of subjects wear multiple wristbands for the same amount of time. The wristbands can then be put through different shipping and storage conditions to confirm if there are any differences, and if so, which conditions are optimal for the study. For example, wristbands could be cut into halves and stored at different temperature conditions for different lengths of time. One wristband could be shipped to a secondary location then back to the lab. The detection frequency and concentrations of compounds from each wristband under different storage and shipping conditions can be compared to determine if there is a significant difference that makes certain of conditions optimal for each study. Field and transport blanks should be collected, especially when shipping at ambient temperatures.

In conclusion, silicone wristbands have promise as a convenient tool for exposure studies. Although sample sizes in current studies are limited, compounds in wristbands appear to correlate well with levels of compounds and metabolites in paired air or biomarker samples, indicating the promise of worn silicone wristbands for exposure assessment. They are easy to deploy and wear and can be transported at ambient temperatures. The wristbands detect complex mixtures of environmental chemicals: for PAHs, studies reported the detection of multiple PAHs in the wristbands from exposures as short as several days in non-occupationally exposed persons. However, many research gaps remain with regards to determining factors affecting the concentrations of chemicals detected in the wristbands and interpretation of the data. The SWB may be especially useful where high absolute precision in exposure assessment is not needed for the purpose of the study; for example they can be useful in an intervention study aiming to reduce pesticide exposure at the group level or in epidemiological studies where participants are grouped into a few exposure categories (e.g., 'high' and 'low' exposure). For these reasons, silicone wristbands can help increase inclusivity and representation of exposure across different populations who may not be able to easily participate in traditional exposure studies, such as children, pregnant women, and farmers and farmworkers, especially those in developing countries or remote areas. It is important to note that some of these populations are often among the most susceptible and most exposed to harmful exposures.

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