Environment International 170 (2022) 107524

Contents lists available at ScienceDirect

Environment International

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

Full length article

Personal air pollutant exposure monitoring in South African children in the VHEMBE birth cohort

Kayley DeLay [a,b,1], Elizabeth Z. Lin [a,1], Jeremy P. Koelmel [a], Riana Bornman [c], Muvhulawa Obida [c], Jonathan Chevrier [d,*], Krystal J. Godri Pollitt [a,b,*]

[a] Department of Environmental Health Sciences, Yale School of Public Health, New Haven, CT 06520, USA
[b] Department of Chemical and Environmental Engineering, Yale School of Engineering and Applied Sciences, New Haven, CT 06520, USA
[c] University of Pretoria Institute for Sustainable Malaria Control and School of Health Systems and Public Health, University of Pretoria, Pretoria, South Africa
[d] Department of Epidemiology, Biostatistics and Occupational Health, Faculty of Medicine and Health Sciences, McGill University, Montréal, QC, Canada

ARTICLE INFO

Handling Editor: Shoji Nakayama

Keywords:
South Africa
Children's health
Passive sampling
Personal monitoring
Biomass burning
Pesticides

ABSTRACT

The burden of disease associated with environmental exposures disproportionately impacts residents of low- and middle-income countries. Children living in rural regions of these countries may experience higher exposure to insecticides from indoor residual spraying used for malaria control and household air pollution. This study evaluated environmental exposures of children living in a rural region of South Africa. Quantifying exposure levels and identifying characteristics that are associated with exposure in this geographic region has been challenging due to limitations with available monitoring techniques. Wearable passive samplers have recently been shown to be a convenient and reliable tool for assessing personal exposures. In this study, a passive sampler wristband, known as Fresh Air wristband, was worn by 49 children (five-years of age) residing in the Limpopo province of South Africa. The study leveraged ongoing research within the Venda Health Examination of Mothers, Babies, and their Environment (VHEMBE) birth cohort. A wide range of chemicals (35 in total) were detected using the wristbands, including polycyclic aromatic hydrocarbons (PAHs), organochlorine pesticides, phthalates, and organophosphate esters (OPEs) flame retardants. Higher concentrations of PAHs were observed among children from households that fell below the food poverty threshold, did not have access to electric cookstoves/burners, or reported longer times of cooking or burning materials during the sampling period. Concentrations of *p,p′*-DDD and *p,p′*-DDT were also found to be elevated for children from households falling below the food poverty threshold as well as for children whose households were sprayed for malaria control within the previous 1.5 years. This study demonstrates the feasibility of using passive sampler wristbands as a non-invasive method for personal exposure assessment of children in rural regions of South Africa to complex mixtures environmental contaminants derived from a combination of sources. Future studies are needed to further identify and understand the effects of airborne environmental contaminants on childhood development and strategies to mitigate exposures.

1. Introduction

Air pollution is ubiquitous, but the burden of disease resulting from toxic exposures disproportionately affects residents of low- and middle-income countries (LMICs) (WHO 2018). Over 600,000 estimated deaths each year result from exposure to air pollution in Africa alone (ACGIH 2018). Children are particularly susceptible to exposure because of their developing airways, immature immune system, as well as higher respiratory rates, food intake per body weight, and skin-to-volume ratios relative to adults (Kim et al., 2018; Masekela and Vanker 2020; Sly and Bush 2015; Voynow and Auten 2015). In rural South Africa, exposures from household air pollution and indoor residual spraying (IRS) are of major concern. The main source of exposure to household air pollution originates from burning solid fuels for cooking (Shezi and Wright 2018). While the transition to cleaner cooking technologies has progressed globally, many regions in Africa, and in other LMICs, still use polluting

* Corresponding authors at: 60 College Street, Room 523, New Haven, CT 06520, USA (K.J. Godri Pollitt). 2001 McGill College Ave, Montréal, QC H3A 1G1, Canada (J. Chevrier).
E-mail addresses: jonathan.chevrier@mcgill.ca (J. Chevrier), krystal.pollitt@yale.edu (K.J. Godri Pollitt).
[1] Equal contribution.

https://doi.org/10.1016/j.envint.2022.107524
Received 21 October 2021; Received in revised form 2 September 2022; Accepted 13 September 2022
Available online 16 September 2022

fuels and technologies (Gordon et al., 2014; World Health, 2018). Over 80% of the African region primarily rely on these polluting energy sources for everyday household activities like cooking, heating, and lighting (WHO 2018). Childhood exposure to household air pollution can lead to both acute and chronic respiratory conditions ranging from increased frequency in cough, wheezing, and acute respiratory tract infections to long-term ailments like chronic obstructive pulmonary disease (Fullerton et al., 2008; Masekela and Vanker 2020). Those living in rural regions of Africa can be exposed to insecticides, such as *p,p'*-dichlorodiphenyltrichloroethane (*p,p*′-DDT), from IRS conducted for malaria control. These chemicals are generally applied to the interior walls and rafters of traditional mud dwellings, and underneath the eaves on the outside of the homes (Bouwman et al., 2006; Gitari et al., 2018). *p,p*′-DDT exposure is of particular concern for early childhood development because of its potential for neurotoxicity and endocrine disruption (Slima et al., 2017; Eskenazi et al., 2018; Liu and Schelar 2012; Oulhote and Bouchard 2013; Patisaul and Adewale 2009; Viel et al., 2017).

Assessing children's exposure to environmental chemicals can be challenging as few non-invasive methods that can be easily deployed on a large scale exist. The use of wearable passive samplers has grown in popularity in the past decade and has shown to be an efficient approach for evaluating personal exposures (Gibson et al., 2019; Hammel et al., 2016; Koelmel et al., 2021; Koelmel et al., 2022; Lin et al., 2020; O'Connell et al., 2014; Wang et al., 2019). These emerging tools are an extension of stationary passive samplers that have been used for the last two decades to collect airborne contaminants with polyurethane foam (PUF) (Gibson et al., 2019; Hammel et al., 2016; D Kim et al., 2018; O'Connell et al., 2014; Wang et al., 2019), styrene divinylbenzene copolymer (XAD) (Wania et al., 2003; Wania and Shunthirasingham 2020), and polyethylene (Messier et al., 2019). Sampling devices that can be worn allow for the evaluation of exposure from both outdoor and indoor spaces as well as of the variability specific to individuals.

Over 40 environmental health studies have evaluated personal exposures using wearable passive samplers with most worn as a wristband (Okeme et al., 2022). Two wristband designs have been reported, including a commercial band and the Fresh Air wristband. The former is a loop of silicone rubber that is retailed by vendors as a 'Livestrong' or awareness bracelet. The silicone is a sorbent for environmental contaminants that are taken up from the air and the skin of the participant capturing a combination of inhalation and dermal exposure pathways. The Fresh Air wristband similarly uses a silicone rubber (polydimethylsiloxane, PDMS) as a sorbent to collect environmental contaminants. The sorbent in the Fresh Air wristband, however, is encased in a perforated chamber mounted in a band to restrict assessment to inhalation exposures. The lightweight, non-invasive features of wristband passive samplers enhance the feasibility of investigating the exposures of vulnerable populations, like pregnant women (Gibson et al., 2019), the older adults (Koelmel et al., 2021), and children (Koelmel et al., 2022; Levasseur et al., 2021; Quintana et al., 2019) compared to active sampling counterparts (Doherty et al., 2021).

Numerous studies in the US have used these wearable tools to characterise personal exposures of organophosphate ester flame retardants (OPFRs) (Hammel et al., 2016; Reddam et al., 2020), polycyclic aromatic hydrocarbons (PAHs) (Dixon et al., 2018; Hendryx et al., 2020; Paulik et al., 2018; Rohlman et al., 2019a; Rohlman et al., 2019b), polybrominated diphenyl ethers (PBDEs) (Hammel et al., 2018), pesticides (Aerts et al., 2018), and a variety of pollutants (Anderson et al., 2017; Bergmann et al., 2018; Craig et al., 2019; Donald et al., 2019; Nicole, 2018; Romanak et al., 2019; Wang et al., 2019; Shaorui Wang et al., 2020; Zuy et al., 2020). Other studies in the US have examined children's exposures to PBDEs (Kile et al., 2016; Lipscomb et al., 2017), OPFRs (Gibson et al., 2019; Hammel et al., 2020; Kile et al., 2016; Lipscomb et al., 2017), phthalates (Hammel et al., 2020), PAHs (Lin et al., 2020), pesticides (Harley et al., 2019; Vidi et al., 2017), and nicotine (Quintana et al., 2019). Most recently, wearable passive air samplers have been used to evaluate personal exposure to airborne particles, including respiratory virus (Angel et al., 2022). There have also been studies using wristbands conducted on adults outside of the US including West Africa (Donald et al., 2016), Peru (Bergmann et al., 2017), Chile (Manzano et al., 2019), Bangladesh (Wang et al., 2020), Japan (Reche et al., 2020), China (Guo et al., 2021; Koelmel et al., 2021), a combination of participants from France and Italy (S. Wang et al., 2020), and a combination of participants from South Africa, Peru, and the US (Dixon et al., 2019). There has been limited study of personal pollutant exposures for children outside of the US, particularly in LMICs. A recent publication assessed the exposure of 38 children (median age of 12 years) and a guardian from the same household that lived in agricultural communities in the southwest region of South Africa (Fuhrimann et al., 2022). Of the 21 pesticides evaluated using silicone wristbands, 16 were detected, including deltamethrin, chlorpyrifos, boscalid, and cypermethrin. Children were found to have elevated exposure levels to deltamethrin, cypermethrin, *p,p*′-DDE, and *p,p*′-DDT compared to their guardians. Beyond pesticides, further investigation is needed evaluating the mixture of environmental chemicals to which children are exposed in LMICs from other emission sources.

The objective of this study was to evaluate the feasibility of using the novel Fresh Air wristband to assess exposures to diverse range of pollutants derived from various sources, including IRS and combustion activities among five-year-old children residing in the Limpopo province of South Africa. The relationship between detected exposures were further investigated to better understand the context in which exposure arises to inform future exposure mitigation interventions.

2. Materials and methods

2.1. Study population and study design

Participants in the current study were recruited from a larger ongoing birth cohort study, the Venda Health Examination of Mothers, Babies and their Environment (VHEMBE). Participants in the VHEMBE study included 752 mother-child pairs who were recruited between 2012 and 2013 when women presented to give birth at Tshilidzini hospital in Limpopo's Vhembe district. Eligibility criteria included: being at least 18 years of age, living in a home that was <20 km from the hospital, in which Tshivenda was the primary language, planning to remain in the study area for at least two years, having contractions more than five minutes apart at the time of recruitment and not having been infected with malaria during pregnancy (Coker et al., 2018). VHEMBE participants have taken part in follow-up assessments at multiple time points, including home-walkthrough visits and comprehensive surveys at one-week post-partum (Rauch et al., 2018) as well as at 1, 2, 3.5, and 5 years of age (Di Lenardo et al., 2020). Given the size of the cohort, recruitment and visits were scheduled over the course of 18 months. At delivery as well as at each follow-up visit, bilingual (English and Tshivenda) staff members conducted a questionnaire-based interview in Tshivenda to collect data on sociodemographic characteristics, health, and exposure determinants.

Among the 639 children who completed an assessment at 5 years of age, 57 children with a scheduled follow-up visit between September 2018 and January 2019 were recruited and enrolled in the current study (Table S1). Children were given a Fresh Air wristband to wear for three days and were requested to keep the wristband on for all of their daily activities, except while swimming or bathing. At the end of the exposure assessment period, the wristbands were collected for chemical analysis. Four (7%) of the 57 deployed wristbands were lost by participants. Of the 53 retrieved wristbands, some evidence of tampering or damage was found for 35 units (61%). Two of these wristbands (3.5%) were excluded from analysis because of significant damage and two additional units (3.5%) were not analyzed due to technical challenges at the data collection phase. In total, 49 (86%) of the wristbands deployed were used in the present study.

At the end of the exposure assessment period, trained staff administered a questionnaire (Five-year Air Sampling questionnaire) to caretakers to collect detailed information specific to the period during which Fresh Air wristbands were worn by the children. Information was collected about the start and end times of each burning episode (whether for cooking or other purposes), which fuels were used, whether fires occurred indoors or outdoors, and whether the child was present. Cooking and burning time durations reported for the entire sampling period were categorised into long (>8 h), medium (4–8 h), short (0–4 h), and none (0 h). Other information was also recorded by the questionnaire such as the road and traffic conditions near their home, proximity to large factories/warehouses or other major sources of pollution and whether a secondary cooking area was used.

Information from two additional questionnaires was extracted and used in the analysis for this study: the Five-Year Follow-up Visit and the One-Week Postpartum Housing Survey. Information collected from the *Five Year Follow-up Visit* questionnaire used in this study includes questions about households characteristics, such as which fuels were used for cooking, heating, and lighting, poverty status, how smoke or steam may escape from the home, when the home was lasted sprayed for malaria control, and if children lived in the same home as was evaluated in the *One-Week Postpartum Housing Survey*. The Five-Year Follow-up Visit complemented the information collected in the Five-Year Air Sampling questionnaire by providing additional information about each participant's household within the 18 months prior to when the Fresh Air wristbands were deployed. The *One-Week Postpartum Housing Survey* was conducted throughout 2012–2013 following initial enrollment in the VHEMBE study and birth of the child. Information collected during this examination allowed for the assessment of possible longitudinal exposures resulting from household sources. The most pertinent information collected and used from this survey asked about the main sources of energy used for cooking indoors and whether the home was equipped with an electric stove or burner. Information from this survey was used only for participants who lived in the same home at the time of the *Five-Year Follow-up Visit* as they did at the *One-Week Postpartum Housing Survey*. Lists of the questions extracted from each of the three questionnaires are included in the SI (Table S2).

Informed consent was obtained from all caregivers prior to participation in the study. This study was approved by the ethics committees of McGill University, the University of Pretoria, the Limpopo Department of Health and Social Development, Tshilidzini Hospital and Yale University.

2.2. Reagents

Methanol and hexane (Optima LC/MS grade), water and toluene (HPLC grade), dichloromethane (HPLC grade), and polydimethylsiloxane were purchased from Fisher Scientific (Hampton, NH, US). Analytical standards were greater than 98% purity, they were purchased from Accustandard (New Haven, CT, US) and SPEX (Metuchen, NJ, US).

2.3. Air sampling

2.3.1. Passive air samplers

Personal chemical exposures were evaluated using the Fresh Air wristband (Lin et al., 2020). PDMS sorbent bars were used to passively collect airborne contaminants. Four replicate bars were housed in a polytetrafluoroethylene (PTFE) chamber that was mounted in a wristband. Participants in this study wore one wristband for three days. Following the exposure assessment period, all PDMS sorbent bars contained in participants' wristbands were cold chain shipped to the Yale School of Public Health for chemical analysis.

The sampling behaviour of the Fresh Air wristband has been previously described (Lin et al., 2020). Briefly, field calibrations were used to determine the uptake rate and capacity of this wearable passive sampler. Semi-volatile organic compounds reported in the current study were found to reach equilibrium after five to 14 days of sampling. Children enrolled in the VHEMBE cohort wore the Fresh Air wristbands for three days to limit sampling of airborne contaminants to the linear uptake regime.

2.3.2. Sample preparation and storage

All laboratory glassware and tools were rinsed with methanol and baked at 75 °C for at least 24 h before use. PDMS sorbent bars (length = 10 mm, thickness = 0.8 mm, inner diameter = 2 mm) were custom fabricated using high-purity polydimethylsiloxane polymer (Dow Corning) and then cleaned in a vacuum oven (2 h at 260 °C) under of 0.1–0.3 L/min flow of high purity nitrogen (99.99%) prior to use. Cleaned PDMS sorbent bars were individually placed in microvial inserts and stored in air-tight 2 mL amber glass vials with PTFE septa caps. Immediately prior to deployment, a field staff of the study team positioned four cleaned PDMS sorbent bars into the PTFE chambers and mounted these chambers into a wristband. PDMS sorbent bars additionally were secured in place using pre-cleaned neodymium magnets. Due to sometimes long travel distances, field staff stored the Fresh Air wristbands in pre-baked glass jars and carried them to the participants' houses for deployment.

The wristband was worn by participants for 2.9 days (SD = 1.0, range: 2 to 5 days) on average. At the end of the exposure assessment period, a field staff collected and stored the Fresh Air wristband in an air-tight container, then transferred it to the local research lab. Immediately after arrival, the field staff removed the PDMS sorbent bars from each sampler and placed them back to the amber glass vials using stainless steel forceps. Samples were stored at 5 °C prior to analysis. Field blanks were collected throughout the study were used to correct for potential contaminations. These PDMS sorbent bars (field blanks) went through all processing steps that samplers went through, including placement in and out of the form factors (sample preparation), storage, and sample introduction, without being placed on participants.

2.3.3. Sample analysis and data processing

All laboratory glassware and tools were rinsed with methanol and baked at 75 °C for at least 24 h before use. All cleaned glassware and tools were stored at 75 °C in the oven until use. Immediately prior to analysis, PDMS sorbent bars were individually rinsed to removed excess particle/dust collected on samples. As part of this rinse procedure, PDMS sorbent bars were transferred to pre-cleaned 2 mL amber vials using stainless-steel tweezers. Optima LC/MS grade water added to each amber vial and shaken for 10 min at 300 RPM. PDMS sorbent bars were dried using in-house nitrogen for 3 s and were then loaded with an internal standard mixture which contained 5′-fluoro-2,3′,4,4′,5-pentabromodiphenyl ether, naphthalene-d8, acenaphthene-d10, phenanthrene-d10, fluoranthene-d10, pyrene-d10, perylene-d12, and 3,3′,4,4′-tetrabromodiphenyl ether. PDMS sorbent bars were placed into pre-cleaned glass autosampler tubes (Gerstel, Linthicum, MD, USA) on a temperature-controlled autosampler tray maintained at 10 °C (MéCour, Groveland, MA, USA). For sample analysis, an autosampler tube was transferred into a thermal desorption unit (TDU; Gerstel, Linthicum, MD, USA). The TDU was initially held at 30 °C for 1.1 min and then ramped at 720 °C per minute to 280 °C (5 min hold) under a flow rate of 350 mL/min of helium gas (99.999%). Extracted analytes were cyrofocused to −90 °C on a 2 mm, glass wool deactivated liner in a cooled injection system (Gerstel, Linthicum, MD, USA) cooled to −90 °C. The transfer line between the TDU and cooled liner was maintained at 250 °C. Analyses were directly transferred to the gas chromatography (GC) column (TG-5SILMS, 30 m × 0.25 mm × 0.25 μm). The carrier gas flow (helium) was set to 1.4 mL/min and the GC oven was held at 70 °C for one minute and then ramped at 7 °C/min to 300 °C. The final temperature was held for 4.0 min for a total run-time of 37.86 min. During the analysis, full-scan electron ionization (EI) mass spectra (m/z 53.4 – 800) was recorded at an acquisition rate of 4 Hz, and at 60,000 resolution on a Q-Exactive Orbitrap mass spectrometer (ThermoFisher, Waltham, MA, USA). Quality control samples and blanks (laboratory and transport) which were run every five samples.

Raw mass spectral data were analysed using TraceFinder 4.1 (Thermo). Blank feature filtering was conducted using field blanks to remove compounds with high of background contamination following previously described methods (Guo et al., 2021). Briefly, the 95th percentile of individual compound in samples that was less than 10 times of the average of field blanks were removed. Batch-wise median normalization was used to account for potential batch effects (Koelmel et al., 2021). Exposures were quantified for 70 compounds using a seven-point calibration curve (Patterson and Sears 2017). Concentrations below the limit of detection (LOD) were replaced by half the LOD (Xia and Wishart 2011). Wristband mass loadings were $\log_2$ transformed (in pg detected for each PDMS bar) and further normalised by the number of days each participant wore the wristband. Exposure levels were converted to a volume basis (pg/m^3) using a generic uptake coefficient (0.43 m^3/day) determined for the Fresh Air wristband.

Quality assurance and quality control details of the described methods have been previously described (Lin et al., 2020; Guo et al., 2021). Briefly, the thermal desorption extraction procedure used in the current study achieved 80 to 90% recovery of volatile and semi-volatile organic compounds, respectively, from the PDMS sorbent bars. Repeatability of measurements was evaluated for individual compounds at low, mid, and high levels. The standard deviation across three replicates was less than 15% at all test levels for most compounds. Quality control samples analysed in each batch varied by 14% on average for all compounds.

2.4. *Statistical analysis*

Correlations across chemical exposures were explored using

Table 1
Characteristics of study participants and their households.

Participant Characteristic			n	(%)	Mean ± **SD**
Sex	Female		27	(55.1%)	
	Male		22	(44.9%)	
Poverty Status	Below the food poverty threshold (R547 per capita per month)	Yes	24	(49.0%)	
		No	21	(42.9%)	
		No Response	4	(8.2%)	
	Below the lower bound poverty threshold (R785 per capita per month)	Yes	32	(65.3%)	
		No	13	(26.5%)	
		No Response	4	(8.2%)	
	Below the upper bound poverty threshold (R1183 per capita per month)	Yes	37	(75.5%)	
		No	8	(16.3%)	
		No Response	4	(8.2%)	
Malaria Control	Home has been sprayed for malaria control since child's last visit (within the previous 1.5 years)	Yes	12	(24.5%)	
		No	37	(75.5%)	
Waste Management	Rubbish burned on property	Yes	44	(89.8%)	
		No	1	(2.0%)	
		No Response	4	(8.2%)	
	Primary cooking area	Indoors	38	(77.6%)	
		Outdoors	11	(22.4%)	
Cooking	Main source of energy used for cooking indoors*	Electric Stove or Burner	26	(53.1%)	
		Wood	17	(34.7%)	
		Paraffin	2	(4.1%)	
		No indoor cooking	3	(6.1%)	
		No Response	1	(2.0%)	
	While the child was at home or around home, papers, cow dung, wood, corn cobs, straw, leaves or other vegetation were burned	Yes	37	(75.5%)	
		No	11	(22.5%)	
		Unknown	1	(2.0%)	
	Average number of separate cooking occurrences				2.29 ± 1.87
	Fuels reported used or burned during sampling period (some were burned in combination)	Wood	91	(70.5%)	
		Papers	18	(14.0%)	
		Straw	5	(3.9%)	
		Leaves	4	(3.1%)	
		Other Vegetation	2	(1.6%)	
		Cow Dung	1	(0.8%)	
		Corn Cobs	0	(0%)	
		Unknown	8	(6.2%)	
	Daily average of the amount of time spent cooking or burning fuels (min)	Total time spent daily			119 ± 114
		Of total, the time spent burning materials only indoors			89 ± 118
		Of total, the time spent burning materials while child was directly near the fire			56 ± 86

Note: Percentage may not total to 100% due to rounding.

Spearman's rank correlations since most of the exposures did not follow a normal distribution after transformation. To compare exposure levels by cooking methods and time, malaria controls, poverty level a Kruskal-Wallis test was performed.

3. Results

3.1. Characteristics of study population

3.1.1. Participant Characteristics

Participant and household characteristics throughout the three-day sampling period are summarised in Table 1. Additional information about the study population is shown in Table S3. A total of 49 participants aged five-years were included in this study, with 55.1% ($n = 27$) being female. Over 75% ($n = 37$) of the participants' households were below the upper bound poverty threshold based on household income (1183 Rand per capita per month; equivalent to about $65 US), 32 of these 37 participants were below the lower bound poverty threshold (785 Rand per capita per month; equivalent to about $44 US), and 24 of these, nearly half (49%) of the study population, fell below the food poverty threshold (547 Rand per capita per month; equivalent to about $30 US).

3.1.2. Household characteristics

The primary area for cooking in participants' homes was an indoor kitchen space (77.6%) with 40.2% ($n = 16$) of these households reporting having a secondary cooking location either indoors ($n = 7$) or outdoors ($n = 9$). Out of all the fires that occurred throughout the sampling period, wood was the fuel most burned, making 70.5% of the total types of fuels reported being used, followed by paper, at 14.0%.

Over three-quarters of the participants' caregivers reported fuels being burned during the sampling period. Time spent cooking/burning during the exposure assessment period ranged from 0 ($n = 11$) to 19 h per day with the daily average of time spent cooking/burning materials being 119 (SD = 114) minutes. While information was not collected about the cooking appliance used in participants' homes at the Five-Year

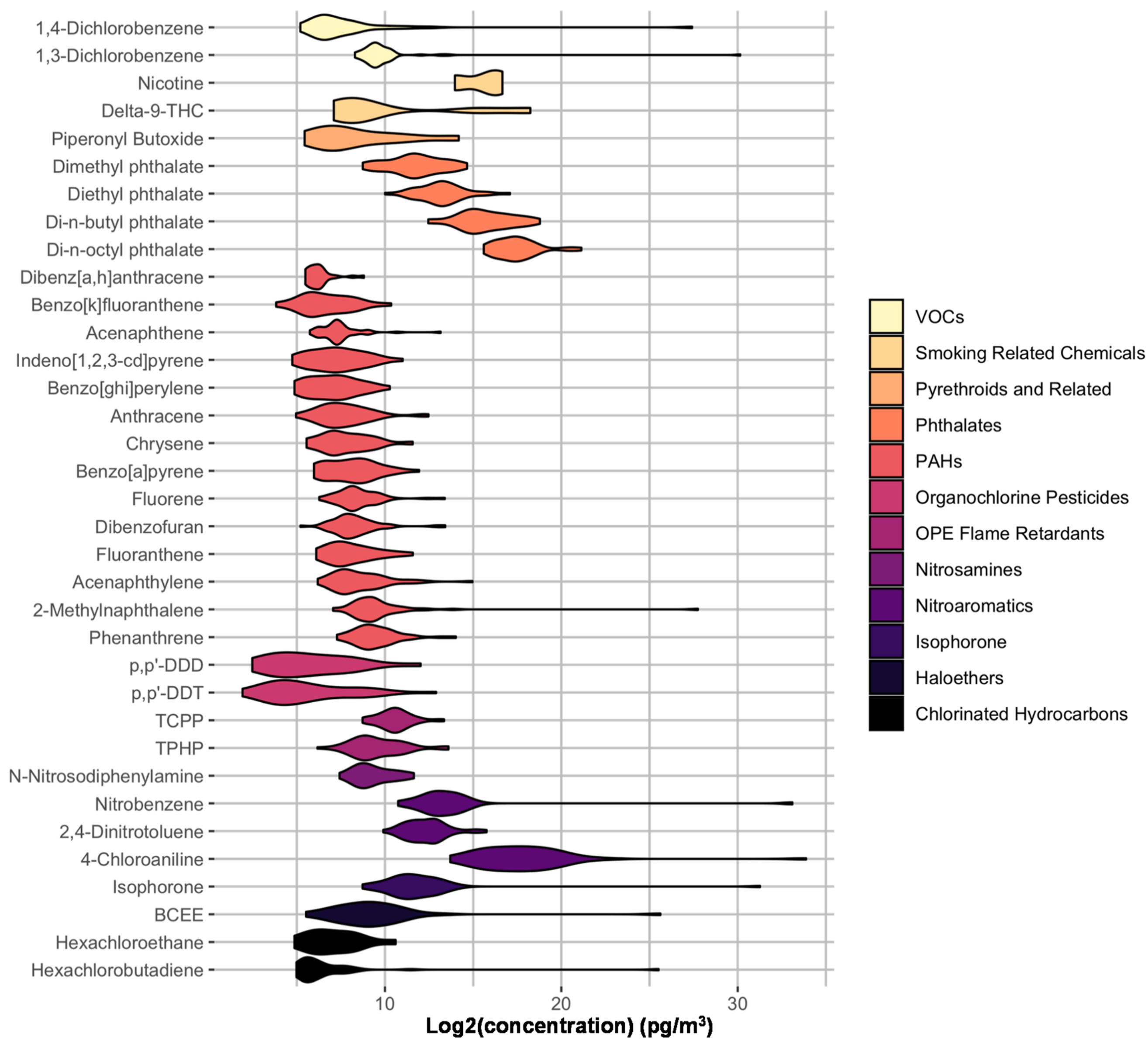

Fig. 1. Summary of the 35 contaminants detected using the Fresh Air wristbands collected from the 5-year-old study population (n = 49). Abbreviations: volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs), organophosphate esters (OPEs), tetrahydrocannabinol (THC), bis(2-chloro-1-methylethyl) ether (BCEE), tris(1-chloro-2-propyl) phosphate (TCPP), triphenyl phosphate (TPHP), *p,p'*-dichlorodiphenyldichloroethane (*p,p'*-DDD), *p,p'*-dichlorodiphenyltrichloroethane (*p,p'*-DDT).

Follow-up Visit, 55.1% of children had not moved since birth when a One-Week Postpartum Housing survey was conducted. From this original survey, 53.1% of participants' households used an electric stove or burner for cooking; others used either wood cookstoves or paraffin cookstoves (38.8%). Nearly 90% of the homes burned waste on their property and about one-quarter (n = 12) of the participants' homes had been sprayed for malaria control within the previous 1.5 years of the survey. Of these 12 households sprayed, seven reported DDT was the pesticide used. All homes were naturally ventilated by opening windows and doors.

3.2. Exposure assessment

Fresh Air wristbands were analysed for 70 compounds, spanning 16 chemical classes. Of these, 35 compounds were not detected in the samples collected in this study, including brominated flame retardants, fungicides, and herbicides (Table S4). The other 35 compounds were detected in at least one of the samplers above the limit of detection and field blanks (Table S5). This included 14 PAHs, four phthalates, three nitroaromatics, two organochlorine pesticides, two organophosphate ester (OPE) flame retardants, two volatile organic compounds (VOCs), two chlorinated hydrocarbon, two smoking-related compounds, a pyrethroid-related compound, a haloether, a nitroamine, and isophorone.

The distribution of detected chemicals is shown in Fig. 1. All four phthalates, di-n-butyl phthalate, di-n-octyl phthalate, diethyl phthalate, dimethyl phthalate, were detected in 100% of the wristbands at higher mass loadings compared to the other chemical families, with medians of 4.42×10^4, 1.74×10^5, 9.35×10^3, and 3.36×10^3 pg/m^3. Two organochlorine pesticides were analysed: *p,p′*-DDT and *p,p'*- dichlorodiphenyldichloroethane (*p,p′*-DDD). *p,p′*-DDT was detected in 77.6% of the wristbands at a median concentration of 27.3 pg/m^3 and *p,p′*-DDD was detected in 44.9% of the wristbands at a median concentration of 42.6 pg/m^3. Multiple PAHs were detected in all participants' wristbands with 2-methylnapthalene having the highest detected mass loading median at 631 pg/m^3 and dibenz[a,h]anthracene with the lowest median at 69.0 pg/m^3. Both OPE flame retardants analysed, TCPP and triphenyl phosphate (TPHP), were detected in 100% of the wristbands with median concentrations of 1.48×10^3 and 553 pg/m^3, respectively. Nitroaromatics and isophorone generally had elevated detection mass loading, particularly 4-chloroaniline (median = 2.08×10^5 pg/m^3; 100% detection frequency). Hexachlorobutadiene (HCBD) was detected in 77.6% wristbands with a median concentration of 62.0 pg/m^3 and hexachloroethane was detected in 26.5% wristbands with a median concentration of 119 pg/m^3. The haloether, *bis*(chloroethyl)ether (BCEE), was detected in 39 of the 49 wristbands with a median concentration of 489 pg/m^3. N-nitrosodiphenylamine exposure was detected for all participants with a median concentration of 529 pg/m^3. Two VOCs, 1,3-dichlorobenzene and 1,4-dichlorobenzene were detected in 100% and 79.6% of the wristbands, respectively. Piperonyl butoxide, a synergist added to pyrethroid insecticide commercial mixtures, was detected in 42.9% of wristbands with a median concentration of 202 pg/m^3. Two smoking-related compounds were detected in the wristbands, nicotine (6.1%) and delta-9-tetrahydrocannabinol (THC-9) (16.3%).

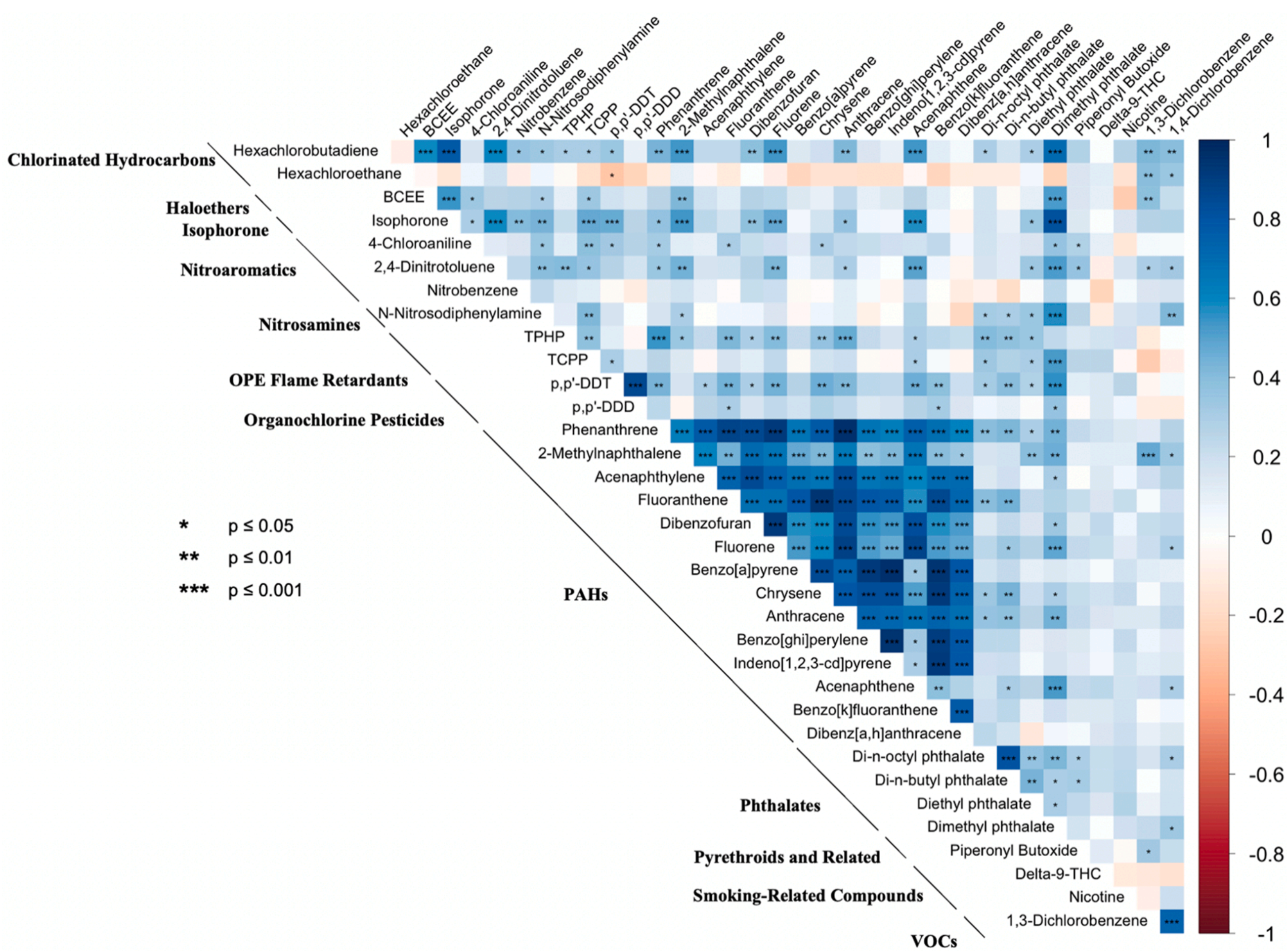

Fig. 2. Correlation matrix of the personal chemical exposures assessed using Fresh Air wristbands, grouped by chemical family.

3.3. Comparison across exposures

Spearman's rank correlations were evaluated across the 35 analytes detected (Fig. 2). Positive relationships were found between PAHs, including phenanthrene and anthracene (ρ = 0.96, p = 1.67 × 10^{-27}), benzo(a)pyrene and indeno[1,2,3-cd] pyrene (ρ = 0.96, p = 1.28 × 10^{-26}), benzo[ghi]perylene and indeno[1,2,3-cd] pyrene (ρ = 0.94, p = 1.96 × 10^{-24}). VOCs were also found to be correlated, including 1–3-dichlorobenzene and 1,4-dichlorobenzene (ρ = 0.73, p = 1.99 × 10^{-9}).

The two organochlorine compounds were further observed to be correlated, *p,p'*-DDT and *p,p'*-DDD (ρ = 0.85, p = 8.45 × 10^{-15}). Other correlations include hexachlorobutadiene and isophorone (ρ = 0.78, p = 5.28 × 10^{-11}), dimethyl phthalate and isophorone (ρ = 0.81, p = 1.18 × 10^{-12}), hexachlorobutadiene and dimethyl phthalate (ρ = 0.70, p = 2.98 × 10^{-8}). Two phthalates, di-n-octyl phthalate and di-n-butyl phthalate, were found to be positively related (ρ = 0.78, p = 4.16 × 10^{-12}).

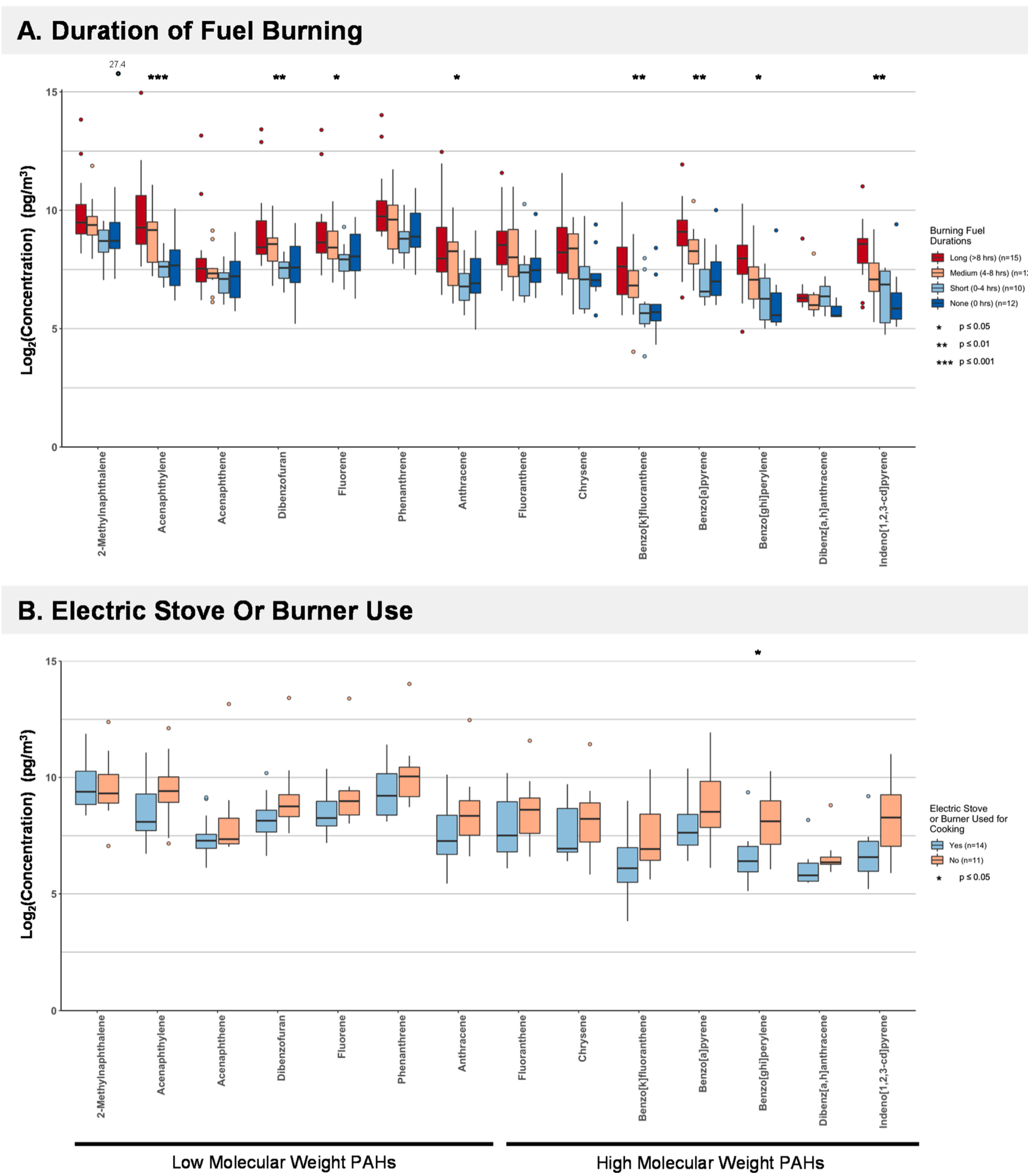

Fig. 3. Personal exposure to PAHs shown by duration of fuel burning (A) and electric stove or burner use (B). Exposures were compared using a Kruskal-Wallis test (p-values ≤ 0.05 and ≤ 0.01 are denoted with asterisks).

3.4. Questionnaire and survey measures

3.4.1. Cooking duration

The amount of time spent cooking or burning materials during the three-day sampling period was recorded in the Five-Year Air Sampling questionnaire. Differences in airborne pollutant concentrations of PAHs were observed based on the durations of time spent cooking or burning materials (Fig. 3A). Increased durations were observed to have generally higher exposure concentrations across PAHs. Differences in concentrations were found for 8 of the 14 compounds in the family ($p \leq 0.05$).), with a $p \leq 0.001$ difference observed for acenaphthylene.

3.4.2. Stove type

Differences in PAH exposure levels were found to correspond to the use of an electric stove or burner for indoor cooking (Fig. 3B). The type of cookstove used by the child's family was recorded in the One-Week Postpartum Housing Survey and analysis was conducted only for those that lived in the same home at the Five-Year Follow-up Visit ($n = 27$). Participants that reported not having an electric stove or burner for cooking were observed to have higher PAH concentrations with significant differences ($p \leq 0.05$) observed for benzo[ghi]perylene.

3.4.3. Poverty status

Evidence of household poverty status being associated with pollutant exposures was observed from wristband concentrations of PAHs and organochlorine pesticides. Higher exposures were found for participants whose household fell below the lower poverty threshold of 785 Rand per capita per month. This trend was apparent across all PAHs, where 9 of the 14 chemicals analysed were found to have a statistically significant difference (Fig. 4A). A similar trend was observed for organochlorine pesticide concentrations. Participants from households below the lower poverty level were observed to have higher concentrations of *p,p′*-DDT (Fig. 4A).

3.4.4. Malaria control

A trend towards higher exposure levels of organochlorine pesticides were observed for participants whose households were sprayed for malaria control within 1.5 years prior to sampling ($n = 12$) (Fig. 4B). In a previous study conducted in the VHEMBE cohort, Gaspar et al., similarly found increased *p,p′*-DDT exposure for mothers that reported having lived in a home sprayed for malaria control (Gaspar et al., 2017). Blood serum concentrations were measured in the mothers at the time of delivery. Concentrations for those from homes that had been sprayed were found to have five to seven times higher levels than those from homes that had never been sprayed.

3.4.5. Ventilation within homes

No association was found between pollutant concentrations and the outlet in which smoke escaped from participant households as reported in the Five-Year Follow-up Visit questionnaire. Households were all naturally ventilated, with smoke escaping from holes in the walls/raised roof, open windows/doors, chimney, or a combination of these outlets. None of the participants reported the use of a kitchen fan with exhaust as a method for ventilation.

3.4.6. Differences by sex

No difference in pollutant concentrations were found between female and male children across all chemical families, except for one phthalate. Females were found to have elevated exposure to di-n-octyl phthalate, compared to male participants ($p \leq 0.05$). This was also the only compound observed to have higher levels in households that reported cooking indoors during the 3-sampling period rather than outdoors.

4. Discussion

Wearable passive samplers have been shown to be an assessable method of capturing personal exposure profiles, with their use being particularly suited for vulnerable populations (Koelmel et al., 2021; Lin et al., 2020). This study is the first to present quantitative exposure levels of children residing in a rural region of South Africa to diverse mixture of environmental contaminants derived from multiple emission sources, including household air pollution and IRS. This study confirmed expected trends with the use of Fresh Air wristbands, such as positive relationship between cooking time and exposure level, and further highlight individual and household characteristics that influence personal exposure levels to select contaminants. We detected enhanced trends in PAH exposures associated with longer reported durations of burning materials during sampling period, lower household poverty status, and those who had previously reported not having access to an

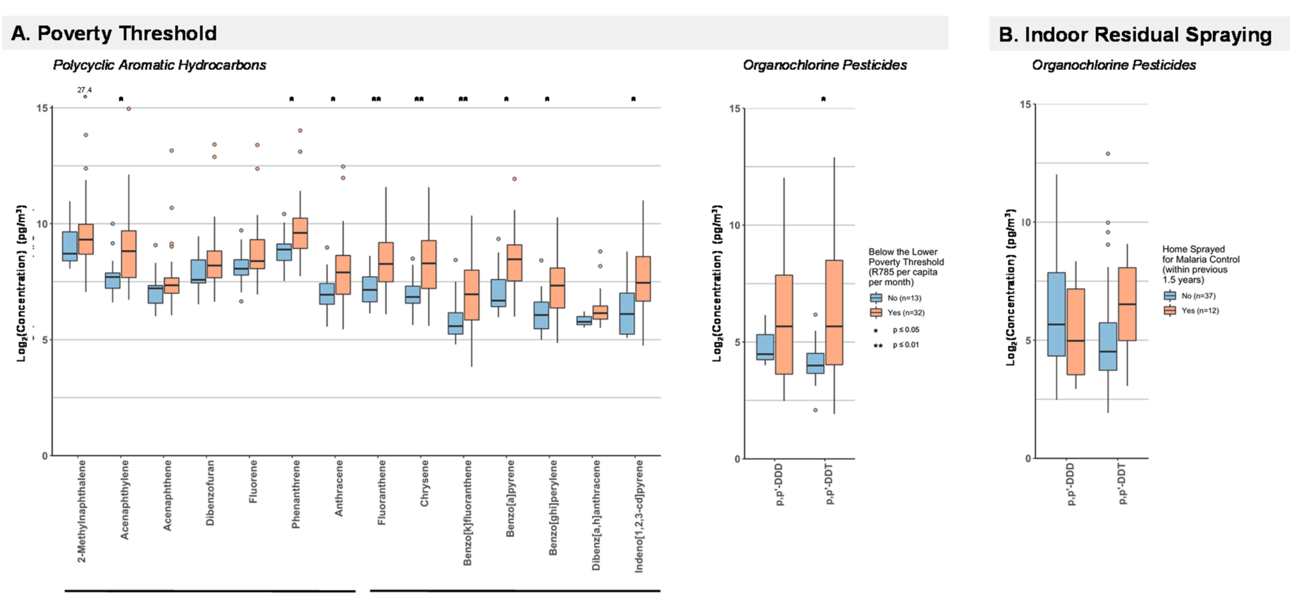

Fig. 4. Personal chemical exposures are shown by poverty status (A) and indoor residual spraying (B). Poverty status was evaluated as households below the lower poverty threshold (R785 per capita per month). Exposures were compared using a Kruskal-Wallis test (p-values ≤ 0.05 and ≤ 0.01 are denoted with asterisks).

electric stove or burner for indoor cooking. We found distinct trends in pesticide concentrations for participant who reported that their homes had been recently (previous 1.5 years) sprayed for malaria control, as well as observed higher concentrations for households that fell below the lower poverty threshold. Our findings provide insights for future policy implications on air pollution issues in rural areas of South Africa.

4.1. PAHs

PAHs comprised nearly half of the contaminants detected in the study participants' wristbands. Of the 14 PAHs evaluated, 13 are designated as high priority PAHs by the United States Environmental Protection Agency (US EPA) given their known toxicity to human health and prevalence in the environment. Five of these are classified by the International Agency for Research on Cancer as either carcinogenic (Group 1), probably carcinogenic (Group 2A), or possibly carcinogenic (Group 2B) to humans: benzo[a]pyrene (1), dibenz[a,h]anthracene (2A), indeno[1,2,3-cd] pyrene (2B), chrysene (2B), and benzo[k]fluoranthene (2B) (IARC 2020).

The magnitude of PAH concentrations detected were comparable across the chemical family, with the highest levels found for phenanthrene and 2-methylnapthalene. The positive correlation identified among PAHs suggests a similar source. These airborne contaminants have been previously attributed to vegetation fires from agricultural practices and residential biomass burning in similar geographic region (Lacaux et al., 1991; Lacaux et al., 1996; Lammel et al., 2013). Burning activities within households are an important source of exposure, as trends with burning fuel durations were identified by increased PAH pollutant concentrations in the study population. Exposure to PAHs results from pyrolysis or incomplete combustion, primarily driven by cooking related emissions (CDC 2022; Pratiti 2021). Over 80% of the African region primarily relies on polluting fuels and technologies for cooking, and utilise these fuels, including coal, wood, charcoal, dung, crop residues, or kerosene, for other daily activities like heating and lighting (WHO 2018). Household air pollution is exacerbated by inadequate ventilation systems in homes, and the use of open fires and traditional stoves, without flues or hoods, is common. Families that lived in the same households for at least five years and reported not having an electric burner or stove available for cooking when they were first surveyed were observed to have higher concentrations of PAHs, specifically of benzo[a]pyrene, benzo[ghi]perylene, benzo[k]fluoranthene, and indeno[1,2,3-cd] pyrene ($p \leq 0.05$). Increased concentrations were particularly evident for children from households falling below the lower poverty threshold of 785 Rand per capita per month, likely due to an increased use of solid fuels, which further raises concerns about long term exposure to elevated environmental pollutant concentrations in some homes within the VHEMBE cohort as compared with others. This finding was supported by increased use of electric stoves by children from families above the poverty threshold (31%) compared to other stove types (0%).

The longer that study participant's household reported cooking or burning materials during the sampling period, the higher PAH concentrations were measured. Various cooking factors influence PAH emissions beyond the duration alone, such as the type of food and cooking style, as well as household features which can influence the movement of air and ventilation of pollutants in the home (Ahmed et al., 2019; Munyeza et al., 2020).

4.2. Organochlorine insecticides

p,p′-DDD and, *p,p′*-DDT, were found at lower concentrations compared to the other 33 airborne contaminants detected using the wristbands; however, any detection of these chemicals is notable given their well-recognised toxicity. DDT is used for malaria control, which is particularly important as the estimated number of worldwide cases of malaria in 2018 was 228 million with approximately 405,000 deaths, where young children account for most of all malaria deaths globally (WHO 2019). In South Africa, 9,540 malaria cases have been reported, resulting in 69 deaths in 2018 (WHO 2019). These cases were primary reported for the provinces of Limpopo, Mpumalanga and KwaZulu-Natal (Munzhedzi et al., 2021). IRS has been found to be effective in mitigating malaria transmission, but potential negative health effects have been associated with exposure to *p,p′*-DDT and its breakdown products (Barlow et al., 2001; Bouwman et al., 2006). Studies have shown that *p,p′*-DDT has a long half-life, estimated between 6 and 10 years, making this insecticide persistent in the environment and able to bioaccumulate in humans (Longnecker 2005; Wolff et al., 2000). DDT is a neurotoxin and endocrine disruptor and of particular concern for early childhood development (Eskenazi et al., 2018; Liu and Schelar 2012; Oulhote and Bouchard 2013; Patisaul and Adewale 2009; Slima et al., 2017; Viel et al., 2017).

In South Africa, IRS with DDT has been used since the 1940s and continues to be a applied to control malaria vectors in Southern Africa despite it being banned by most countries (Eskenazi et al., 2018; Sharp and le Sueur 1996). DDT is typically applied to interior walls and roofs of traditional unpainted mud dwellings (Bouwman et al., 2006; Gaspar et al., 2017; Gitari et al., 2018; Verner et al., 2018). Participants in this study who were from homes reportedly sprayed within the previous 1.5-years and who were from households that fell below the lower poverty threshold had higher wristband DDT concentrations. These results suggest that IRS applications used for malaria control are associated with higher exposures, similar to findings from other studies using more invasive serum concentration measurements (Gaspar et al., 2017; Whitworth et al., 2014). Furthermore, previous studies focused on adult exposure. This is one of the first studies showing airborne exposure concentrations for children. Verner et al., measured serum concentrations of *p,p′*-DDT and *p,p′*-DDE in children within the VHEMBE cohort at 12 and 24 months of age and found higher serum levels among children relative to their mothers' at the time of at delivery (Verner et al., 2018).

4.3. Phthalates

The four detected phthalates, di-n-butyl phthalate, di-n-octyl phthalate, diethyl phthalate, dimethyl phthalate, are commonly used as plasticisers and used to help retain color and fragrance in personal care products (Lyche 2011). Phthalates are suspected endocrine disruptors and childhood exposures have been linked to increased risk of allergic diseases, such as asthma and eczema, autism, hyperactivity, and neurodegenerative diseases (Jurewicz and Hanke 2011; Kim et al., 2018). Childhood exposure is of particular concern due to the incidence of higher exposures compared to adults, in combination with developmental effect (Braun et al., 2013; Gani et al., 2021; Hammel et al., 2020).

The high molecular weight phthalate, di-n-octyl phthalate, was more abundant than the other three lower molecular weight compounds. Higher molecular weight phthalates are prevalent in indoor environments, with uses in food packaging and in building materials, and often exist in the particle phase and partially in the gas phase due to higher boiling points and lower volatility (Hammel et al., 2020; Wei et al., 2018; Zhou et al., 2021). Di-n-octyl phthalate was the only pollutant observed to be higher in concentration for participants of households that reported cooking primarily indoors during the sampling period rather than outdoors. Females were found to have higher concentrations of di-n-octyl phthalate than males. Previous studies in Africa have shown that women and girls tend to have elevated exposures of criteria airborne pollutants ($PM_{2.5}$, PM_{10}, CO) than men, typically reflecting an overall increased time spent indoors and distinctions in household activities including cooking (Bruce et al., 2000; Ezzati et al., 2000; Okello et al., 2018). More frequent use of personal care products among women compared to men may also lead to increased exposures (Zota and Shamasunder 2017). This trend was not notably observed for pollutants measured in our study population, aside from the single analyte, di-n-octyl phthalate, nor were phthalates measured in these previous studies.

4.4. Other airborne contaminants

The detected haloether, BCEE, was found in most wristband samples at elevated concentrations. The compound is primarily produced for use in pesticides and the production of other chemicals. There is limited research regarding the toxicity of BCEE, but some evidence suggests this compound may lead to respiratory irritation, impact the nervous system, and body weight (ATSDR 2017). Children in this study may have inhaled BCEE released from industrial activity in the region. BCEE has not been previously detected in similar cohorts.

HCBD was also frequently detected in participants' wristbands. This chemical is typically produced as a byproduct in the manufacturing of other chlorinated hydrocarbons and can be used or produced in the production of solvents, lubricants, heat transfer liquid and hydraulic liquid, and pesticides (UN 2012; USEPA 2003). Few studies have assessed inhalation exposure of HCBD (Balmer et al., 2019; Rauert et al., 2018; Wong et al., 2021). While the health effects of HCBD in humans are unclear, studies have shown HCBD to be toxic for the kidney in rats (Green et al., 2003; Lock and Ishmael 1979). Other evidence from animal studies suggests HCBD may be linked to irritation, nervous system depression, and damage to fatty liver degeneration (UN 2016). The US EPA has classified HCBD as a potential carcinogen and it was added as a persistent organic pollutant under the Stockholm Convention in 2015 (UN 2015; USEPA 2003).

Other detected contaminants of concern include N-nitrosodiphenylamine, 1,4-dichlorobenzene, 4-chloroaniline, 2,4-dinitrotoluene, nitrobenzene, and isophorone. Aside from a study comparing personal VOC concentrations for women living in rural small holder dairy farms which found that 1,4-dichlorobenzene concentrations were lower for women who used biogas for fuel for cooking, rather than solid biomass, these airborne contaminants have not been measured in rural populations in Africa (Dohoo et al., 2015).

4.5. Limitations and future directions

Fresh Air wristbands facilitate comprehensive exposure assessment to environmental contaminants at different developmental stages given their non-invasive wearable design. Questions, however, remain regarding the comparability of this emerging technology with traditional exposure assessment tools in various sampling conditions and is the subject of ongoing studies. This study conducted targeted analysis of 70 contaminants collected by the wristbands. While this assessment provides broad coverage of airborne exposures, this panel of contaminants represents only a fraction of environmental factors to which participants may be exposed through inhalation, ingestion, and dermal pathways. Exposure to these chemicals was evaluated over three days, which we acknowledge provides only a snapshot of children's environments. Compared to other assessment approaches, such as measurement of exposure markers in blood which offer insights into longer-term exposure trends for persistent chemicals, the three-day monitoring period is indicative of more recent exposures. Exposures have also been commonly evaluated using urine samples. It is important to recognize that urinary markers are representative of exposures over an even shorter window (∼24 h). As part of this study, information collected from questionnaires was used to interpret results. The children that participated in this study resided in rural regions of South Africa with a high prevalence of malaria. The practice of IRS for infectious disease control in this area limits generalizability of results to children living in other parts of the country.

Exposure assessment of participants in the VHEMBE cohort using the Fresh Air wristbands is ongoing with the aim of understanding the role of environmental contaminants on child growth and development, and to develop strategies to mitigate these exposures. Based on results from the current study, additional questions were added to the VHEMBE questionnaires regarding "trash" and "plastic" burning. We plan to evaluate environmental exposures longitudinally as additional wristband samples are collected from children. Expanded targeted analysis to include other compounds of interest, such as *p,p*-DDE and other pyrethroids, are planned as part of future studies, as well as non-targeted chemical analyses of wristband samplers.

5. Conclusions

Wearable passive samplers enable widescale evaluation of airborne pollutant exposures and are particularly suited for vulnerable populations, such as children. This study is the first of its kind, evaluating exposures using passive sampling techniques for children in rural Africa to assess exposures from known sources such as IRS and biomass burning. The findings indicate exposure to a variety of airborne pollutants with suspected adverse health effects including organochlorine pesticides used for malaria control (DDT), PAHs resulting from combustion, and phthalates. Trends associated with higher levels of exposure to select analytes were identified in the study population: longer durations of cooking or burning materials, lower poverty households, homes without previous access to an electric stove or burner, and households that were sprayed for malaria control. Findings demonstrate the utility of the Fresh Air wristband at highlighting differences in households that may contribute to increased exposures. Understanding trends that lead to increased exposure will advance in the development of strategies to mitigate such exposures. Exposure to toxic environmental contaminants suggest a need for further evaluation of children's pollutant exposures in South Africa and other rural areas in low- and middle-income countries.

CRediT authorship contribution statement

Kayley DeLay: Conceptualization, Data curation, Investigation, Methodology, Formal analysis, Validation, Visualization, Writing – original draft. **Elizabeth Z. Lin:** Conceptualization, Investigation, Data curation, Methodology, Formal analysis, Validation, Writing – original draft. **Jeremy P. Koelmel:** Conceptualization, Data curation, Formal analysis, Investigation. **Riana Bornman:** Methodology, Investigation, Data curation, Resources. **Muvhulawa Obida:** Investigation, Data curation, Project administration, Resources. **Jonathan Chevrier:** Conceptualization, Data curation, Funding acquisition, Investigation, Methodology, Project administration, Resources, Writing – review & editing. **Krystal J. Godri Pollitt:** Conceptualization, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Visualization, Writing – original draft.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was supported by the Canadian Institute for Health Research and a Canada Research Chair in Global Environmental Health and Epidemiology (JC).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2022.107524.

References

ACGIH, 2018. Documentation of the threshold limit values and biological exposure indices, 7th ediition - 2018 supplement. Cincinnati, OH:American Conference of Governmental Industrial Hygienists.

Aerts, R., Joly, L., Szternfeld, P., Tsilikas, K., De Cremer, K., Castelain, P., Aerts, J.-M., Van Orshoven, J., Somers, B., Hendrickx, M., Andjelkovic, M., Van Nieuwenhuyse, A.n., 2018. Silicone wristband passive samplers yield highly individualized pesticide residue exposure profiles. Environ. Sci. Technol. 52 (1), 298–307.

Ahmed, F., Hossain, S., Hossain, S., Fakhruddin, A.N.M., Abdullah, A.T.M., Chowdhury, M.A.Z., Gan, S.H., 2019. Impact of household air pollution on human health: Source identification and systematic management approach. SN Appl. Sci. 1 (5).

Anderson, K.A., Points, G.L., Donald, C.E., Dixon, H.M., Scott, R.P., Wilson, G., Tidwell, L.G., Hoffman, P.D., Herbstman, J.B., O'Connell, S.G., 2017. Preparation and performance features of wristband samplers and considerations for chemical exposure assessment. J. Eposure Sci. Environ. Epidemiol. 27 (6), 551–559.

Angel, D.M., Gao, D., DeLay, K., Lin, E.Z., Eldred, J., Arnold, W., Santiago, R., Redlich, C., Martinello, R.A., Sherman, J.D., Peccia, J., Godri Pollitt, K.J., 2022. Development and application of a polydimethylsiloxane-based passive air sampler to assess personal exposure to sars-cov-2. Environ. Sci. Technol. Lett. 9 (2), 153–159.

ATSDR, 2017. Toxicological profile for bis(2-chloroethyl) ether (bcee). Atlanta, GA.

Balmer, J.E., Hung, H., Vorkamp, K., Letcher, R.J., Muir, D.C.G., 2019. Hexachlorobutadiene (hcbd) contamination in the arctic environment: A review. Emerging Contaminants 5, 116–122.

Barlow, S.M., Sullivan, F.M., Lines, J., 2001. Risk assessment of the use of deltamethrin on bednets for the prevention of malaria. Food Chem. Toxicol. 39 (5), 407–422.

Bergmann, A.J., North, P.E., Vasquez, L., Bello, H., del Carmen Gastañaga Ruiz, M., Anderson, K.A., 2017. Multi-class chemical exposure in rural peru using silicone wristbands. J. Eposure Sci. Environ. Epidemiol. 27 (6), 560–568.

Bergmann, A.J., Points, G.L., Scott, R.P., Wilson, G., Anderson, K.A., 2018. Development of quantitative screen for 1550 chemicals with gc-ms. Anal. Bioanal. Chem. 410 (13), 3101–3110.

Bouwman, H., Sereda, B., Meinhardt, H.M., 2006. Simultaneous presence of ddt and pyrethroid residues in human breast milk from a malaria endemic area in south africa. Environ. Pollut. 144 (3), 902–917.

Braun, J.M., Sathyanarayana, S., Hauser, R., 2013. Phthalate exposure and children's health. Curr. Opin. Pediatr. 25, 247–254.

Bruce, N., Perez-Padilla, R., Albalak, R., 2000. Indoor air pollution in developing countries: A major environmental and public health challenge. Bull World Health Organ 78, 1078–1092.

CDC. 2022. Polycyclic aromatic hydrocarbons (pahs) factsheet. Available: https://www.cdc.gov/biomonitoring/PAHs_FactSheet.html [accessed March 15, 2022].

Coker, E., Chevrier, J., Rauch, S., Bradman, A., Obida, M., Crause, M., Bornman, R., Eskenazi, B., 2018. Association between prenatal exposure to multiple insecticides and child body weight and body composition in the vhembe south african birth cohort. Environ. Int. 113, 122–132.

Craig, J.A., Ceballos, D.M., Fruh, V., Petropoulos, Z.E., Allen, J.G., Calafat, A.M., Ospina, M., Stapleton, H.M., Hammel, S., Gray, R., Webster, T.F., 2019. Exposure of nail salon workers to phthalates, di(2-ethylhexyl) terephthalate, and organophosphate esters: A pilot study. Environ. Sci. Technol. 53 (24), 14630–14637.

Di Lenardo, T.Z., Ward, B.J., Pillet, S., Mann, K., Bornman, R., Obida, M., et al., 2020. Exposure to lead and vaccine-specific igg titers in south african children participating in the venda health examination of mothers, babies and their environment (vhembe): A longitudinal study. Environ. Res. 180, 108794.

Dixon, H.M., Scott, R.P., Holmes, D., Calero, L., Kincl, L.D., Waters, K.M., Camann, D.E., Calafat, A.M., Herbstman, J.B., Anderson, K.A., 2018. Silicone wristbands compared with traditional polycyclic aromatic hydrocarbon exposure assessment methods. Anal. Bioanal. Chem. 410 (13), 3059–3071.

Dixon, H.M., Armstrong, G., Barton, M., Bergmann, A.J., Bondy, M., Halbleib, M.L., Hamilton, W., Haynes, E., Herbstman, J., Hoffman, P., Jepson, P., Kile, M.L., Kincl, L., Laurienti, P.J., North, P., Paulik, L.B., Petrosino, J., Points, G.L., Poutasse, C.M., Rohlman, D., Scott, R.P., Smith, B., Tidwell, L.G., Walker, C., Waters, K.M., Anderson, K.A., 2019. Discovery of common chemical exposures across three continents using silicone wristbands. R. Soc. Open Sci. 6 (2).

Doherty, B.T., Koelmel, J.P., Lin, E.Z., Romano, M.E., Godri Pollitt, K.J., 2021. Use of exposomic methods incorporating sensors in environmental epidemiology. Curr. Environ. Health Reports 8 (1), 34–41.

Dohoo, C., Read Guernsey, J., Gibson, M.D., VanLeeuwen, J., 2015. Impact of biogas digesters on cookhouse volatile organic compound exposure for rural kenyan farmwomen. J. Expo Sci. Environ. Epidemiol. 25 (2), 167–174.

Donald, C.E., Scott, R.P., Blaustein, K.L., Halbleib, M.L., Sarr, M., Jepson, P.C., Anderson, K.A., 2016. Silicone wristbands detect individuals' pesticide exposures in west africa. R. Soc. Open Sci. 3 (8).

Donald, C.E., Scott, R.P., Wilson, G., Hoffman, P.D., Anderson, K.A., 2019. Artificial turf: Chemical flux and development of silicone wristband partitioning coefficients. Air Qual. Atmos. Health 12 (5), 597–611.

Eskenazi, B., An, S., Rauch, S.A., Coker, E.S., Maphula, A., Obida, M., Crause, M., Kogut, K.R., Bornman, R., Chevrier, J., 2018. Prenatal exposure to ddt and pyrethroids for malaria control and child neurodevelopment: The vhembe cohort, south africa. Environ. Health Perspect. 126 (4).

Ezzati, M., Saleh, H., Kammen, D.M., 2000. The contributions of emissions and spatial microenvironments to exposure to indoor air pollution from biomass combustion in kenya. Environ. Health Perspect. 108 (9), 833–839.

Fuhrimann, S., Mol, H.G.J., Dias, J., Dalvie, M.A., Röösli, M., Degrendele, C., Figueiredo, D.M., Huss, A., Portengen, L., Vermeulen, R., 2022. Quantitative assessment of multiple pesticides in silicone wristbands of children/guardian pairs living in agricultural areas in south africa. Sci. Total Environ. 812.

Fullerton, D.G., Bruce, N., Gordon, S.B., 2008. Indoor air pollution from biomass fuel smoke is a major health concern in the developing world. Trans R Soc Trop Med Hyg 102 (9), 843–851.

Gani, K.M., Hlongwa, N., Abunama, T., Kumari, S., Bux, F., 2021. Emerging contaminants in south african water environment- a critical review of their occurrence, sources and ecotoxicological risks. Chemosphere 269, 128737.

Gaspar, F.W., Chevrier, J., Quirós-Alcalá, L., Lipsitt, J.M., Barr, D.B., Holland, N., Bornman, R., Eskenazi, B., 2017. Levels and determinants of ddt and dde exposure in the vhembe cohort. Environ. Health Perspect. 125 (7), 077006.

Gibson, E.A., Stapleton, H.M., Calero, L., Holmes, D., Burke, K., Martinez, R., Cortes, B., Nematollahi, A., Evans, D., Anderson, K.A., Herbstman, J.B., 2019. Differential exposure to organophosphate flame retardants in mother-child pairs. Chemosphere 219, 567–573.

Gitari W, Makoni T, Somerset V, Onaneye-Babajide O. 2018. Spatial distribution of ddt and its metabolites in soils from indoor residual spraying in tshilamusi village, limpopo.

Gordon, S.B., Bruce, N.G., Grigg, J., Hibberd, P.L., Kurmi, O.P., Lam, K.-B., Mortimer, K., Asante, K.P., Balakrishnan, K., Balmes, J., Bar-Zeev, N., Bates, M.N., Breysse, P.N., Buist, S., Chen, Z., Havens, D., Jack, D., Jindal, S., Kan, H., Mehta, S., Moschovis, P., Naeher, L., Patel, A., Perez-Padilla, R., Pope, D., Rylance, J., Semple, S., Martin, W. J., 2014. Respiratory risks from household air pollution in low and middle income countries. Lancet Respir. Med. 2 (10), 823–860.

Green, T., Lee, R., Farrar, D., Hill, J., 2003. Assessing the health risks following environmental exposure to hexachlorobutadiene. Toxicol. Lett. 138 (1-2), 63–73.

Guo, P., Lin, E.Z., Koelmel, J.P., Ding, E., Gao, Y., Deng, F., Dong, H., Liu, Y., Cha, Yu'e, Fang, J., Shi, X., Tang, S., Godri Pollitt, K.J., 2021. Exploring personal chemical exposures in china with wearable air pollutant monitors: A repeated-measure study in healthy older adults in jinan, china. Environ. Int. 156.

Hammel, S.C., Hoffman, K., Webster, T.F., Anderson, K.A., Stapleton, H.M., 2016. Measuring personal exposure to organophosphate flame retardants using silicone wristbands and hand wipes. Environ. Sci. Technol. 50 (8), 4483–4491.

Hammel, S.C., Phillips, A.L., Hoffman, K., Stapleton, H.M., 2018. Evaluating the use of silicone wristbands to measure personal exposure to brominated flame retardants. Environ. Sci. Technol. 52, 11875–11885.

Hammel, S.C., Hoffman, K., Phillips, A.L., Levasseur, J.L., Lorenzo, A.M., Webster, T.F., Stapleton, H.M., 2020. Comparing the use of silicone wristbands, hand wipes, and dust to evaluate children's exposure to flame retardants and plasticizers. Environ. Sci. Technol. 54 (7), 4484–4494.

Harley, K.G., Parra, K.L., Camacho, J., Bradman, A., Nolan, J.E.S., Lessard, C., Anderson, K.A., Poutasse, C.M., Scott, R.P., Lazaro, G., Cardoso, E., Gallardo, D., Gunier, R.B., 2019. Determinants of pesticide concentrations in silicone wristbands worn by latina adolescent girls in a california farmworker community: The cosecha youth participatory action study. Sci. Total Environ. 652, 1022–1029.

Hendryx, M., Wang, S., Romanak, K.A., Salamova, A., Venier, M., 2020. Personal exposure to polycyclic aromatic hydrocarbons in appalachian mining communities. Environ. Pollut. 257.

IARC, 2020. Agents classified by the iarc monographs. Available: https://monographs.iarc.who.int/agents-classified-by-the-iarc/ [accessed May 1, 2022].

Jurewicz, J., Hanke, W., 2011. Exposure to phthalates: Reproductive outcome and children health. A review of epidemiological studies. Int. J. Occup. Med. Environ. Health 24, 115–141.

Kile, M.L., Scott, R.P., O'Connell, S.G., Lipscomb, S., MacDonald, M., McClelland, M., Anderson, K.A., 2016. Using silicone wristbands to evaluate preschool children's exposure to flame retardants. Environ. Res. 147, 365–372.

Kim, D., Chen, Z.i., Zhou, L.-F., Huang, S.-X., 2018a. Air pollutants and early origins of respiratory diseases. Chronic Diseases Translational Med. 4 (2), 75–94.

Kim, Y.-M., Kim, J., Cheong, H.-K., Jeon, B.-H., Ahn, K., Meliker, J., 2018b. Exposure to phthalates aggravates pulmonary function and airway inflammation in asthmatic children. PLoS ONE 13 (12), e0208553.

Koelmel, J.P., Lin, E.Z., Guo, P., Zhou, J., He, J., Chen, A., Gao, Y., Deng, F., Dong, H., Liu, Y., Cha, Yu'e, Fang, J., Beecher, C., Shi, X., Tang, S., Godri Pollitt, K.J., 2021. Exploring the external exposome using wearable passive samplers - the china bape study. Environ. Pollut. 270.

Koelmel, J.P., Lin, E.Z., DeLay, K., Williams, A.J., Zhou, Y., Bornman, R., Obida, M., Chevrier, J., Godri Pollitt, K.J., 2022. Assessing the external exposome using wearable passive samplers and high-resolution mass spectrometry among south african children participating in the vhembe study. Environ. Sci. Technol. 56 (4), 2191–2203.

Lacaux, J.P., Lefeivre, B., Delmas, R.A., Cros, B., Andreae, M.O., 1991. Influence of biomass burning emissions on precipitation chemistry in the equatorial forests of africa. United States:Massachusetts Inst of Tech Press.

Lacaux, J.P., Delmas, R., Jambert, C., Kuhlbusch, T.A.J., 1996. Nox emissions from african savanna fires. Journal of Geophysical Research: Atmospheres 101 (D19), 23585–23595.

Lammel, G., Heil, A., Stemmler, I., Dvorská, A., Klánová, J., 2013. On the contribution of biomass burning to pops (pahs and pcdds) in air in africa. Environ. Sci. Technol. 47 (20), 11616–11624.

Levasseur, J.L., Hammel, S.C., Hoffman, K., Phillips, A.L., Zhang, S., Ye, X., Calafat, A.M., Webster, T.F., Stapleton, H.M., 2021. Young children's exposure to phenols in the home: Associations between house dust, hand wipes, silicone wristbands, and urinary biomarkers. Environ. Int. 147.

Lin, E.Z., Esenther, S., Mascelloni, M., Irfan, F., Godri Pollitt, K.J., 2020. The fresh air wristband: A wearable air pollutant sampler. Environ. Sci. Technol. Lett. 7 (5), 308–314.

Lipscomb, S.T., McClelland, M.M., MacDonald, M., Cardenas, A., Anderson, K.A., Kile, M. L., 2017. Cross-sectional study of social behaviors in preschool children and exposure to flame retardants. Environ. Health 16, 23.
Liu, J., Schelar, E., 2012. Pesticide exposure and child neurodevelopment: Summary and implications. Workplace Health Saf 60 (5), 235–242.
Lock, E.A., Ishmael, J., 1979. The acute toxic effects of hexachloro-1: 3-butadiene on the rat kidney. Arch. Toxicol. 43 (1), 47–57.
Longnecker, M.P., 2005. Invited commentary: Why ddt matters now. Am. J. Epidemiol. 162, 726–728.
Lyche, J.L., 2011. Chapter 48 - phthalates. In: Reproductive and developmental toxicology, (Gupta RC, ed). San Diego:Academic Press, 637-655.
Manzano, C.A., Dodder, N.G., Hoh, E., Morales, R., 2019. Patterns of personal exposure to urban pollutants using personal passive samplers and gc × gc/tof–ms. Environ. Sci. Technol. 53 (2), 614–624.
Masekela, R., Vanker, A., 2020. Lung health in children in sub-saharan africa: Addressing the need for cleaner air. Int. J. Environ. Res. Public Health 17, 6178.
Messier, K.P., Tidwell, L.G., Ghetu, C.C., Rohlman, D., Scott, R.P., Bramer, L.M., Dixon, H.M., Waters, K.M., Anderson, K.A., 2019. Indoor versus outdoor air quality during wildfires. Environ. Sci. Technol. Lett. 6 (12), 696–701.
Munyeza, C.F., Osano, A.M., Maghanga, J.K., Forbes, P.B.C., 2020. Polycyclic aromatic hydrocarbon gaseous emissions from household cooking devices: A kenyan case study. Environ. Toxicol. Chem. 39 (3), 538–547.
Munzhedzi, M., Rogawski McQuade, E.T., Guler, J.L., Shifflett, P.E., Krivacsy, S., Dillingham, R., et al., 2021. Community knowledge, attitudes and practices towards malaria in ha-lambani, limpopo province, south africa: A cross-sectional household survey. Malar. J. 20, 188.
Nicole, W., 2018. Wristbands for Research: Using Wearable Sensors to Collect Exposure Data after Hurricane Harvey. Environ. Health Perspect. 126 (4), 042001.
O'Connell, S.G., Kincl, L.D., Anderson, K.A., 2014. Silicone wristbands as personal passive samplers. Environ. Sci. Technol. 48 (6), 3327–3335.
Okello, G., Devereux, G., Semple, S., 2018. Women and girls in resource poor countries experience much greater exposure to household air pollutants than men: Results from uganda and ethiopia. Environ. Int. 119, 429–437.
Okeme, J.O., Koelmel, J.P., Johnson, E., Lin, E.Z., Gao, D., Godri Pollit, K.J., 2022. Wearable passive samplers for assessing environmental exposure to organic chemicals: Current approaches and future directions Current Environmental Health Reports, in review.
Oulhote, Y., Bouchard, M.F., 2013. Urinary metabolites of organophosphate and pyrethroid pesticides and behavioral problems in canadian children. Environmental Health Perspectives 121, 1378-1384.
Patisaul, H, Adewale, H., 2009. Long-term effects of environmental endocrine disruptors on reproductive physiology and behavior. Frontiers in Behavioral Neuroscience 3.
Patterson, R.E., Sears, D.D., 2017. Metabolic effects of intermittent fasting. Annu. Rev. Nutr. 37 (1), 371–393.
Paulik, L.B., Hobbie, K.A., Rohlman, D., Smith, B.W., Scott, R.P., Kincl, L., Haynes, E.N., Anderson, K.A., 2018. Environmental and individual pah exposures near rural natural gas extraction. Environ. Pollut. 241, 397–405.
Pratiti, R., 2021. Household air pollution related to biomass cook stove emissions and its interaction with improved cookstoves. AIMS Public Health 8, 309–321.
Quintana, P.J.E., Hoh, E., Dodder, N.G., Matt, G.E., Zakarian, J.M., Anderson, K.A., Akins, B., Chu, L., Hovell, M.F., 2019. Nicotine levels in silicone wristband samplers worn by children exposed to secondhand smoke and electronic cigarette vapor are highly correlated with child's urinary cotinine. J. Eposure Sci. Environ. Epidemiol. 29 (6), 733–741.
Rauch, S., Bradman, A., Coker, E., Chevrier, J., An, S., Bornman, R., Eskenazi, B., 2018. Determinants of exposure to pyrethroid insecticides in the vhembe cohort, south africa. Environ. Sci. Technol. 52 (21), 12108–12121.
Rauert, C., Harner, T., Schuster, J.K., Eng, A., Fillmann, G., Castillo, L.E., Fentanes, O., Ibarra, M.V., Miglioranza, K.S.B., Rivadeneira, I.M., Pozo, K., Aristizábal Zuluaga, B. H., 2018. Air monitoring of new and legacy pops in the group of latin america and caribbean (grulac) region. Environ. Pollut. 243, 1252–1262.
Reche, C., Viana, M., van Drooge, B.L., Fernández, F.J., Escribano, M., Castaño-Vinyals, G., Nieuwenhuijsen, M., Adami, P.E., Bermon, S., 2020. Athletes' exposure to air pollution during world athletics relays: A pilot study. Sci. Total Environ. 717.
Reddam, A., Tait, G., Herkert, N., Hammel, S.C., Stapleton, H.M., Volz, D.C., 2020. Longer commutes are associated with increased human exposure to tris(1,3-dichloro-2-propyl) phosphate. Environ. Int. 136, 105499.
Rohlman, D., Dixon, H.M., Kincl, L., Larkin, A., Evoy, R., Barton, M., et al., 2019a. Development of an environmental health tool linking chemical exposures, physical location and lung function. BMC Public Health 19, 854.
Rohlman, D., Donatuto, J., Heidt, M., Barton, M., Campbell, L., Anderson, K., Kile, M., 2019b. A case study describing a community-engaged approach for evaluating polycyclic aromatic hydrocarbon exposure in a native american community. Int. J. Environ. Res. Public Health 16 (3), 327.
Romanak, K.A., Wang, S., Stubbings, W.A., Hendryx, M., Venier, M., Salamova, A., 2019. Analysis of brominated and chlorinated flame retardants, organophosphate esters, and polycyclic aromatic hydrocarbons in silicone wristbands used as personal passive samplers. J. Chromatogr. A 1588, 41–47.
Sharp, B.L., le Sueur, D., 1996. Malaria in south africa–the past, the present and selected implications for the future. S Afr Med J 86, 83–89.
Shezi, B., Wright, C.Y., 2018. Household air pollution exposure and respiratory health outcomes: A narrative review update of the south african epidemiological evidence. Clean Air Journal 28, 43–56.
Slima, B.A., Chtourou, Y., Barkallah, M., Fetoui, H., Boudawara, T., Gdoura, R., 2017. Endocrine disrupting potential and reproductive dysfunction in male mice exposed to deltamethrin. Hum. Exp. Toxicol. 36, 218–226.
Sly, P.D., Bush, A., 2015. From the cradle to the grave: The early-life origins of chronic obstructive pulmonary disease. Am. J. Respir. Crit. Care Med. 193, 1–2.
UN. Risk profile on hexachlorobutadiene, persistent organic pollutants review committee (poprc), unep/pops/poprc.8/16/add.2. In: Proceedings of the Stockholm Convention on Persistent Organic Pollutants, 2012, United Nations Environment Programme.
UN. Report of the conference of the parties to the stockholm convention on persistent organic pollutants on the work of its seventh meeting, unep/pops/cop.7/36*. In: Proceedings of the Stockholm Convention on Persistent Organic Pollutants, 2015, United Nations Environment Programme.
UN. Report of the persistent organic pollutants review committee on the work of its twelfth meeting, unep/pops/poprc.12/11/add.5. In: Proceedings of the Stockholm Convention on Persistent Organic Pollutants, 2016, United Nations Environment Programme.
USEPA. 2003. Candidate contaminant list regulatory determination support document for hexachlorobutadiene. EPA-815-R-03-11. Washington, DC.
Verner, M.-A., Chevrier, J., Ngueta, G., Rauch, S., Bornman, R., Eskenazi, B., 2018. Early-life exposure to p, p′-ddt and p, p′-dde in south african children participating in the vhembe study: An assessment using repeated serum measurements and pharmacokinetic modeling. Environ. Int. 119, 478–484.
Vidi, P.-A., Anderson, K.A., Chen, H., Anderson, R., Salvador-Moreno, N., Mora, D.C., et al., 2017. Personal samplers of bioavailable pesticides integrated with a hair follicle assay of DNA damage to assess environmental exposures and their associated risks in children. Mutation Research/Genetic Toxicology and Environmental Mutagenesis 822, 27–33.
Viel, J.-F., Rouget, F., Warembourg, C., Monfort, C., Limon, G., Cordier, S., et al., 2017. Behavioural disorders in 6-year-old children and pyrethroid insecticide exposure: The pelagie mother–child cohort. Occup. Environ. Med. 74, 275–281.
Voynow, J.A., Auten, R., 2015. Environmental pollution and the developing lung. Clin. Pulm. Med. 22, 177–184.
Wang, S., Romanak, K.A., Stubbings, W.A., Arrandale, V.H., Hendryx, M., Diamond, M. L., et al., 2019. Silicone wristbands integrate dermal and inhalation exposures to semi-volatile organic compounds (svocs). Environ. Int. 132, 105104.
Wang, S., Romanak, K.A., Hendryx, M., Salamova, A., Venier, M., 2020a. Association between thyroid function and exposures to brominated and organophosphate flame retardants in rural central appalachia. Environ. Sci. Technol. 54, 325–334.
Wang, S., Romanak, K.A., Tarallo, S., Francavilla, A., Viviani, M., Vineis, P., et al., 2020b. The use of silicone wristbands to evaluate personal exposure to semi-volatile organic chemicals (svocs) in france and italy. Environ. Pollut. 267, 115490.
Wang, Y., Peris, A., Rifat, M.R., Ahmed, S.I., Aich, N., Nguyen, L.V., et al., 2020c. Measuring exposure of e-waste dismantlers in dhaka bangladesh to organophosphate esters and halogenated flame retardants using silicone wristbands and t-shirts. Sci. Total Environ. 720, 137480.
Wania, F., Shen, L., Lei, Y.D., Teixeira, C., Muir, D.C.G., 2003. Development and calibration of a resin-based passive sampling system for monitoring persistent organic pollutants in the atmosphere. Environ. Sci. Technol. 37, 1352–1359.
Wania, F., Shunthirasingham, C., 2020. Passive air sampling for semi-volatile organic chemicals. Environ Sci Process Impacts 22, 1925–2002.
Wei, W., Mandin, C., Ramalho, O., 2018. Influence of indoor environmental factors on mass transfer parameters and concentrations of semi-volatile organic compounds. Chemosphere 195, 223–235.
Whitworth, K.W., Bornman, R.M.S., Archer, J.I., Kudumu, M.O., Travlos, G.S., Wilson, R. E., et al., 2014. Predictors of plasma ddt and dde concentrations among women exposed to indoor residual spraying for malaria control in the south african study of women and babies (sowb). Environ. Health Perspect. 122, 545–552.
WHO, 2018. Air pollution and child health: Prescribing clean air: Summary. Geneva, Switzerland:World Health Organization.
WHO. 2019. World malaria report 2019. 9789241565721. Geneva, Switzerland:World Health Organization.
Wolff, M.S., Zeleniuch-Jacquotte, A., Dubin, N., Toniolo, P., 2000. Risk of breast cancer and organochlorine exposure. Cancer Epidemiol. Biomark. Prev. 9, 271–277.
Wong, F., Hung, H., Dryfhout-Clark, H., Aas, W., Bohlin-Nizzetto, P., Breivik, K., et al., 2021. Time trends of persistent organic pollutants (pops) and chemicals of emerging arctic concern (ceac) in arctic air from 25 years of monitoring. Sci. Total Environ. 775, 145109.
World Health O, 2018. Air pollution and child health: Prescribing clean air: Summary. Geneva:World Health Organization.
Xia, J., Wishart, D.S., 2011. Web-based inference of biological patterns, functions and pathways from metabolomic data using metaboanalyst. Nat. Protoc. 6, 743–760.
Zhou, X., Lian, J., Cheng, Y., Wang, X., 2021. The gas/particle partitioning behavior of phthalate esters in indoor environment: Effects of temperature and humidity. Environ. Res. 194, 110681.
Zota, A.R., Shamasunder, B., 2017. The environmental injustice of beauty: Framing chemical exposures from beauty products as a health disparities concern. Am. J. Obstet. Gynecol. 217, 418.e411–418.e416.
Zuy, Y., Sweck, S.O., Dockery, C.R., Potts, G.E., 2020. Hplc detection of organic gunshot residues collected with silicone wristbands. Anal. Methods 12, 85–90.