Indoor Air Exposure to Multiple Agricultural Pesticides Potentially Posing the Highest Risk to Young Children

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ABSTRACT

Pesticides are extensively used to improve crop yield and quality in many African countries where agricultural production is a critical economic activity, including Eswatini (formerly Swaziland) in southern Africa. However, the potential health effects of indoor exposure to agricultural pesticides are deeply concerning, and children are particularly vulnerable. This case study assessed such exposure and its risks by sampling and analyzing the air inside the homes of 15 pesticide applicators and 12 non-applicators in Swazi communities surrounded by nearby (~200 m) sugarcane fields on pesticide spraying days. Applying liquid chromatography with tandem mass spectrometry (LC-MS/MS) to the samples revealed mean ametryn, atrazine, pendimethalin and 2,4-dichlorophenoxyacetic acid (2,4-D) concentrations of 0.75, 0.32, 0.57 and 0.004 $\mu\text{g m}^{-3}$, respectively, in the applicator households and corresponding values of 0.19, 0.03, 0.04 and 0.003 $\mu\text{g m}^{-3}$, respectively, in the non-applicator households. Notably, the non-applicator households exhibited pesticide concentrations far exceeding previously published data. Furthermore, we measured significantly higher levels of ametryn and pendimethalin inside the homes of the applicators than those of the non-applicators. Daily inhalation exposure to agricultural pesticides resulted in cumulative hazard index values above 1.0 at the 95th percentile for children 3 years of age and younger in the applicator households. Thus, the take-home pathway, spray drift and household-to-field distance all play crucial roles in determining the pesticide exposure and health risks indoors. Owing to the dominance of the first factor, we advise applicators to shower and change into clean clothes before returning home to their families. This safety measure is essential for the many families living near agricultural land throughout Africa.

Keywords: Pesticides, Indoor air, Inhalation exposure, Health risk, Eswatini

1 INTRODUCTION

Agricultural production is a very important economic activity in African countries, where pesticides are widely used to maintain high crop yields and quality, improve agricultural productivity and farmers' incomes. However, the application of pesticides potentially leads to indoor exposure amongst people living in agricultural communities, due to spray drift and the take-home pathway

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(Weppner *et al.*, 2006; Chang *et al.*, 2014; Gibbs *et al.*, 2017; Taylor *et al.*, 2020). Residential indoor exposure to pesticides has been associated with deleterious health effects, particularly among women and children, including an increase in risk for breast cancer, menstrual cycle disturbances, adverse birth outcomes, thyroid disorders, leukemia, non-Hodgkin lymphoma, asthma, childhood hematopoietic malignancies, autism spectrum disorders and neurodevelopmental delays following prenatal and early childhood exposure to pesticides (Alavanja *et al.*, 1999; Ma *et al.*, 2002; Windham *et al.*, 2002; Farr *et al.*, 2004; Menegaux *et al.*, 2006; Rudant *et al.*, 2007; El-Zaemey *et al.*, 2013; Shelton *et al.*, 2014; Campos and Freire, 2016; Larsen *et al.*, 2017; Mostafalou and Abdollahi, 2017; Cherry *et al.*, 2018; Hyland *et al.*, 2018; Van Maele-Fabry *et al.*, 2019; Dereumeaux *et al.*, 2020; Chetty-Mhlana *et al.*, 2021). Thus, it is critically important to assess household exposures to pesticides and corresponding health risks for residents living in and around agricultural communities. Families in surrounding residential areas may also potentially be exposed to agricultural pesticides, which can drift from the intended application sites on spraying days.

Several studies have found that close proximity of households to agricultural land is associated with high levels of biological markers of pesticides (Wu *et al.*, 2013; Chevrier *et al.*, 2014; Suarez-Lopez *et al.*, 2018) and high pesticide concentrations in households (Lu *et al.*, 2000; Gibbs *et al.*, 2017). Conversely, other research has not found this positive relationship between household proximity and pesticide levels (Curwin *et al.*, 2005; Ward *et al.*, 2006). Evidence has also shown that pesticides sprayed on fields can drift from the spraying area by up to 400 m (Fenske *et al.*, 2002; Koch *et al.*, 2002; Siebers *et al.*, 2003; Quandt *et al.*, 2004; Carlsen *et al.*, 2006; Wolters *et al.*, 2008) and 750 m (Ward *et al.*, 2006). Moreover, living with a farmworker has been associated with an increase in pesticide concentrations in the household (Gibbs *et al.*, 2017; Bennett *et al.*, 2019; López-Gálvez *et al.*, 2019; Dereumeaux *et al.*, 2020). Research has shown that farmworkers transfer pesticide residues home on their clothes, skin and footwear, and thus expose their family members (Bradman *et al.*, 2011; Coronado *et al.*, 2011b). The presence of agricultural pesticides indoors potentially exposes population groups who are highly sensitive to pesticides, such as children and the elderly (NRI, 1993; Ginsberg *et al.*, 2004). Children are an extremely vulnerable group, because of their greater body weight to body surface area and respiratory, metabolic and circulatory rates compared with adult values (Berglund *et al.*, 1992; Grandjean and Landrigan, 2006; Landrigan and Goldman, 2011; Thompson *et al.*, 2014), enabling pesticide residues to accumulate and distribute substantially faster throughout children's bodies compared with adult bodies (Thompson *et al.*, 2014). The impact of household pesticide exposure in children is therefore of enormous concern.

Household exposure to agricultural pesticides has rarely been studied in African countries and may exhibit different patterns of exposure and levels of susceptibility compared to those in other parts of the world (El-Gamal *et al.*, 2017; Joubert *et al.*, 2019), due to the fact that many farmworkers reside near farm fields. Since 90% of global malaria cases occur in the African region (WHO, 2016), most studies conducted in African countries have focused on the use of indoor residual spraying of dichlorodiphenyltrichloroethane (DDT) and its breakdown product dichlorodiphenyltrichloroethylene (DDE), since DDT is used to control malaria vectors in African countries (Barnhoorn *et al.*, 2009; Van Dyk *et al.*, 2010; Naudé and Rohwer, 2012; Hlongwana *et al.*, 2013; Gaspar *et al.*, 2015; Coker *et al.*, 2018; Murray *et al.*, 2018). To assess agricultural pesticide exposure and associated health risks among farmers and residents in agricultural communities, some studies have relied heavily on questionnaires alone (Oesterlund *et al.*, 2014; da Silva *et al.*, 2016; Manyilizu *et al.*, 2016; Sankoh *et al.*, 2016; Debela *et al.*, 2019), while other African studies have only investigated the presence and concentrations of pesticides in ambient air (Shunthirasingham *et al.*, 2010a, b; Moussaoui *et al.*, 2012; Dalvie *et al.*, 2014; Arinaitwe *et al.*, 2016; Fuhrmann *et al.*, 2020). To the best of our knowledge, this case study is the first to be performed in Africa that has assessed the potential health risks resulting from inhalation exposure to indoor air concentrations of multiple agricultural pesticides sprayed on nearby fields. We collected indoor air samples from applicator and non-applicator households in Eswatini (formerly Swaziland) in southern Africa residing in close proximity (~200 m) to sugarcane fields and analyzed all of the samples for evidence of the most commonly used pesticides in sugarcane production in Eswatini: ametryn, atrazine, pendimethalin and 2,4-dichlorophenoxyacetic acid (2,4-D). We compared airborne levels of these pesticides between applicator and non-applicator households and assessed health risks relating to indoor inhalation exposure of these pesticides. This study sought to highlight the potential impacts of agricultural

pesticides to families and especially children living in agricultural communities in African countries, where many farmworker families may be located very close to farmlands.

2 MATERIALS AND METHODS

2.1 Sampling Area

Sampling was conducted in an agricultural community in Vuvulane, in the north-eastern part of Eswatini. This community resides in a commercial sugarcane farm, situated at 26.07°S and 31.87°E in the Lowveld region of the country. The farm grows sugarcane throughout the year and relies on irrigation systems. For the purpose of this study, households in the community were categorized as either *applicator* or *non-applicator households*. *Applicator households* were defined as households having one or more pesticide applicator(s) living in the household. *Non-applicator households* had no applicator(s) living in the household and members of these households were non-farm workers, whose occupations included teaching in the local community school and factory work. The applicator households belonged to the participants of our previous investigation that assessed the applicators' pesticide inhalation exposures (Msibi *et al.*, 2021). Some of the participants in that study kindly invited us to their homes after completing spraying activities for the day. Neighboring non-applicator households were used as controls. Fig. 1 presents a map of the sampling area. At each household visit, we briefed the members about our sampling procedure and study, in order to allay any concerns or suspicions about the motives of our research. Informed consent was obtained from all households in this study, which received ethical approval from the National Health Research Review Board (NHRRB) in Eswatini.

2.2 Indoor Air Sampling

Indoor air sampling was conducted in 15 applicator and 12 non-applicator households in the afternoons, following the application of pesticides in the fields. To assess indoor pesticide levels, air samples were collected from each residence over 2 consecutive days during the summertime pesticide spraying season in December 2018. The United States Environmental Protection Agency (U.S. EPA) *Compendium of Methods for the Determination of Toxic Organic Compounds in Ambient Air: Second Edition* was used as a guideline (U.S. EPA, 1999). An air sampling apparatus was set up each afternoon to collect air samples in the living rooms of applicators' and non-applicators' homes. In each case, the sampling pump (AirChek 52, Part No. 224–52; SKC, Inc., USA) was mounted on a stand and positioned at a breathing zone height of ~1.4 m. The flow rate was set at 1 L min⁻¹ and XAD-2 OVS tubes (Cat. No. 226-58) were used to capture pesticide residues in the air. The set-up was the same in both sets of households. All applicator and non-applicator households were located within 200 m of the sugarcane fields (Fig. 1).

Fig. 1. A map of Africa showing the location of Eswatini (left), a map of Eswatini (center) and an insert of the sampling area, showing the households surrounded by sugarcane fields (right).

Air temperatures and relative humidity (RH) values were monitored at the start and end of sample collection (Table S1) using a Lutron temperature/humidity monitor (Taipei, Taiwan). We calibrated the sampling pumps on site before and after sample collection, using a Gilibrator-2 Calibrator (Sensidyne, Florida, USA). Variation in the pump flow before and after sampling had to be less than 5%. After sampling, all the relevant data were recorded and sample tubes were securely covered with caps on both ends and sealed in labeled polyethylene zipper bags. Field blanks were also prepared in the living room by using clean, unused cartridges. They were handled, labeled and stored the same way as the collected air samples, except that no air was drawn through them. All samples were stored in a cooler box with ice and transported to the University of Eswatini in Kwaluseni, where they were stored at -10°C in a laboratory, before shipment in dry ice to Taiwan, where they were stored in a -20°C freezer until analysis.

2.3 Pesticide Analysis and Quality Control

LC grade methanol and acetonitrile were purchased from Merck (Darmstadt, Germany). Sigma-Aldrich provided analytical grade standards of ametryn (CAS No. 834-12-8), atrazine (CAS No. 1912-249), pendimethalin (CAS No. 40487-42-1) and 2,4-D (CAS No. 94-75-7), with individual purities exceeding 98% for all chemicals. Sigma-Aldrich also supplied formic acid and ammonium acetate. Cambridge Isotope Laboratories, Inc., supplied the internal standard, atrazine (ethylamine- d_5) $100\text{ }\mu\text{g mL}^{-1}$ in Nonane. The analyte stock solution (1 mg mL^{-1}) and internal standard ($1\text{ }\mu\text{g mL}^{-1}$) were both prepared in acetonitrile, then stored for later dilutions and mixtures in glass vials at -10°C .

Samples were analyzed in June 2019, in Tainan, Taiwan. Liquid chromatography with tandem mass spectrometry (LC-MS/MS) analysis was performed using a Shimadzu UHPLC LC-30A system with an LCMS-8045 triple quadrupole mass spectrometer equipped with a heated electrospray ionization (ESI) probe (Shimadzu Corp., Japan). The instrument was injected with $5\text{ }\mu\text{L}$ of the sample for analysis. Chromatographic separation was achieved using a $100\text{ mm} \times 2.1\text{ mm}$ reverse-phase $2.6\text{ }\mu\text{m}$ SpeedCore C18 column (Fortis Technologies, UK) set at 35°C . A gradient mobile-phase system consisting of Mobiles A and B was used, which worked at a flow rate of $300\text{ }\mu\text{L min}^{-1}$. Mobile A contained 0.4 g ammonium acetate and 1 mL of formic acid in $1,000\text{ mL}$ of ultrapure water, while Mobile B contained 0.4 g ammonium acetate in $1,000\text{ mL}$ of methanol. Due to the differing ESI polarities in the analytes, the mass spectrometer was operated using the multiple reaction monitoring (MRM) mode, incorporating both positive and negative ESI modes, as per our previous paper (Msibi *et al.*, 2021).

For sample extraction, 1.2 mL of methanol was added to the samples. The analytes were then extracted from the samples using an ultrasound bath (Delta Ultrasonic Cleaner; Power Stop Co., Ltd., Bangkok, Thailand) using an ultrasound frequency of 40 kHz and 600 W of power for 30 min . The extracts were removed from the vial using a syringe and a needle, filtered through a nylon filter of $0.22\text{ }\mu\text{m}$ (13 mm diameter) into 1.5 mL vials, capped and labeled. Samples were then run through the LC-MS/MS instrument. The instrument was calibrated for each target compound, using an eight-point calibration curve ranging from 1 to $200\text{ }\mu\text{g L}^{-1}$. Internal standard, atrazine- d_5 ($5\text{ }\mu\text{L}$ of $1\text{ }\mu\text{g mL}^{-1}$), was added to all air samples to quantify the concentrations of the target compounds. Quality assurance/quality control (QA/QC) samples were incorporated during sampling to enhance sample integrity, increase confidence in the analytical data and to check for any contamination. For quality control during instrument analysis, we ran a calibration recovery standard after every 10 samples. Recovery was determined in this study for each target compound. XAD-2 OVS resin was spiked with three different concentrations of the standard: 5 , 50 and $200\text{ }\mu\text{g L}^{-1}$. The spiked resin was left at 4°C for 24 h . Samples were extracted using the sample treatment procedure outlined in this study. The percentage recovery was calculated by comparing instrument analysis results with the concentration spiked into the sample (Ho *et al.*, 2012; Hamsan *et al.*, 2017).

Calibration curves were obtained for all analytes to determine pesticide amounts in indoor air. Eight-point calibration curves were derived from 1 – $200\text{ }\mu\text{g L}^{-1}$, with regression coefficients (R^2) ranging from 0.9965 to 0.9984 . Recovery efficiencies for the spiked XAD-2 OVS resin samples were 100.4% for ametryn, 118.8% for atrazine, 83.8% for pendimethalin and 76.6% for 2,4-D (Table S2).

2.4 Pesticide Concentrations in Households

We used the internal standard to quantify the analyte concentration of indoor air samples. We manually integrated the peak areas of analytes in each sample using the internal standard quantification method. After peak integration, analyte masses were converted to concentrations ($\mu\text{g m}^{-3}$), using stepwise calculations. We initially divided the detected instrument results for each target compound by the recovery for the respective compounds. The results (in $\mu\text{g L}^{-1}$) were then multiplied with 1.2 mL (volume used in extraction) and divided by the volume of sampled air, which was calculated from the sample flow rate and duration. The airborne concentration was calculated using the following equation (HSE, 2015):

$$C_{\text{air}} (\mu\text{g m}^{-3}) = \frac{M \times V}{V_{\text{air}}} \quad (1)$$

In this equation, C_{air} is the concentration of pesticides in air, M is the total mass of analyte in the sample ($\mu\text{g L}^{-1}$), V is the final volume of the extract (0.0012 L) and V_{air} is the volume of air sampled (m^3). V_{air} was calculated using this equation:

$$V_{\text{air}} (\text{m}^3) = \frac{F \times T}{1,000} \quad (2)$$

In this equation, F is the flow rate (L min^{-1}), T is the sampling time (min) and 1,000 is used to convert from L to m^3 . For calculating averages, we assigned a proxy value of half of the limit of detection (LOD), in cases where sample concentrations were below the LOD ($< \text{LOD}$).

2.5 Probabilistic Assessment of Indoor Exposure and Health Risk

Indoor household exposures to pesticides can occur through inhalation, dermal absorption and ingestion pathways (NRI, 2000; Özkara *et al.*, 2016). In this study, the health risk from chronic inhalation exposure of pesticide residues in households was assessed for different age groups (< 0.5 years, 0.5–0.9 years, 1–3 years, 4–12 years, 13–18 years, 19–65 years, > 65 years). Based on the cancer classification of pesticides by the U.S. EPA, all our target compounds were non-carcinogenic (U.S. EPA, 2018) (Table S3). We therefore applied the following equation to calculate the health risk (U.S. EPA, 1997):

$$ADD_i = \frac{C_i \times \text{InhR} \times EF \times ED}{BW \times AT} \quad (3)$$

where $i = 1-4$, representing ametryn, atrazine, pendimethalin and 2,4-D; ADD_i is the average daily dose ($\text{mg kg}^{-1} \text{day}^{-1}$) for i pesticide; C_i is the concentration of i pesticide in the air (mg m^{-3}); InhR is the daily inhalation rate for each age group ($\text{m}^3 \text{day}^{-1}$) (U.S. EPA, 2011; EFSA, 2014); EF is the exposure frequency (260 days y^{-1}); and ED is the exposure duration (years). BW is the body weight (kg) and AT is the averaging time (days). The long-term non-carcinogenic health risk, expressed as the hazard quotient (HQ), was calculated by comparing the inhalation exposure (ADD) with the reference dose (RfD) of each target compound, as follows (U.S. EPA, 1998):

$$HQ_i = \frac{ADD_i}{\text{RfD}_i} \quad (4)$$

The level of concern for the HQ_i value was set as 1, so an HQ_i value > 1 indicates a potential risk. The RfD_i was chosen as the health-based reference value (HBRV) for i pesticide and was cited from the USEPA databases. Cumulative exposures to pesticides were estimated using a hazard index (HI) using the following formula (U.S. EPA, 1998):

$$HI = \sum_{i=1}^4 HQ_i \quad (5)$$

The HI level of concern was set at 1.0; thus, an HI value > 1 indicates that a potential risk may be present.

For probabilistic assessment, we used Microsoft Excel™ 2016 and Oracle® Crystal Ball Release 11.1.2.4.00 software (Oracle Corp., USA) to run 50,000 Monte Carlo (MC) simulations. Prior to estimation of the ADD_i , we used Crystal Ball software to determine the distribution (lognormal) of the concentrations of each pesticide. The inhalation rates and body weight values were also assumed to be lognormally distributed (Table S4) and the correlation coefficient between inhalation rate and body weight was set at 0.5 (U.S. EPA, 2011). Inhalation rate values for the different age groups were obtained from the U.S. EPA *Exposure Factors Handbook: 2011 Edition* (U.S. EPA, 2011). Body weights for adults were adopted from the Eswatini Government's Disease Risk Factor Surveillance Report (DRFSR) (Eswatini Government, 2014) and those for other age groups were adopted from the USEPA *Exposure Factors Handbook*. We estimated the EF to be 260 days y^{-1} divided by 365 days $y^{-1} = 0.712$. Since our measurements were performed on application days, we used the estimated annual number of application days (260 days) as the exposure frequency. This is based on information provided by the applicators. Our study population was separated into age groups, with different body weights and inhalation rates; therefore, we calculated annual risks. The ED was set at 1 year and the AT (days) was calculated as the ED multiplied by 365 days y^{-1} . Thus, ED and AT are essentially simplified to 1. After calculating the ADD_i values for all age groups, we divided them by the RfD_i values for the target compounds to obtain the HQ_i values. The RfD_i values used were 0.009 for ametryn, 0.035 for atrazine, 0.04 for pendimethalin and 0.01 $mg\ kg^{-1}\ day^{-1}$ for 2,4-D (U.S. EPA, 2020) (Table S3).

2.6 Statistical Analysis

Exposure data were handled using Microsoft Excel, to determine descriptive statistics such as frequencies, means and ranges. Some graphs were plotted using R Studio. To determine any significant differences, we compared the concentrations of each pesticide in applicator and non-applicator households using a two-sample *t*-test, assuming unequal variances ($\alpha = 0.05$). Between-group differences were treated as significant when *p*-values were < 0.05 .

3 RESULTS AND DISCUSSION

3.1 Airborne Pesticide Levels in Households

Indoor air samples were collected from 27 households, 15 of which were applicator households and 12 were non-applicator households. Analyte samples in the gas phase were collected in the living rooms of the households, with an average sampling time of 8.99 h (SD = 2.45). Fig. S1 shows a chromatogram of the target compounds from an applicator's household. Fig. 2 shows the concentrations from applicator and non-applicator households. All target compounds except for 2,4-D were detected in all of the samples; detection frequencies for 2,4-D were 33.3% in the applicator and non-applicator households (Table 1). For all target compounds, pesticide concentrations were higher in samples collected from applicators' households compared with those from non-applicator households (Fig. 2); ametryn was the most commonly detected compound in both sets of households, with mean concentrations of $0.75 \pm 1.09\ \mu g\ m^{-3}$ and $0.19 \pm 0.28\ \mu g\ m^{-3}$, respectively. This is consistent with the results of our previous paper (Msibi *et al.*, 2021), which showed that in comparison with the other target compounds, the amount of ametryn used was the highest. The general trend in application rate ($kg\ ha^{-1}$) of the four herbicides was: ametryn $>$ pendimethalin $>$ atrazine $>$ 2,4-D. Our analysis showed that differences were only statistically significant for ametryn ($p = 0.038$) and pendimethalin ($p = 0.03$); the difference between the concentrations from the two sets of households was not statistically significant for atrazine ($p = 0.06$) (see Table 1). Concentrations of pesticides detected in each household are given in Table S5.

3.1.1 Spray drift

In non-applicator households, our data reveal mean indoor air concentrations of $0.19 \pm 0.28\ \mu g\ m^{-3}$ for ametryn, $0.03 \pm 0.03\ \mu g\ m^{-3}$ for atrazine, $0.04 \pm 0.04\ \mu g\ m^{-3}$ for pendimethalin and $0.003 \pm 0.001\ \mu g\ m^{-3}$ for 2,4-D (Table 1). Households in this study were in close proximity ($\sim 200\ m$)

Fig. 2. Box plots showing pesticide concentrations in indoor air samples collected from applicator and non-applicator households. * Statistically significant ($p < 0.05$).

Table 1. Mean concentrations of target compounds from applicator and non-applicator households ($\mu\text{g m}^{-3}$).

Target compounds	Applicator households (n = 15)			Non-applicator households (n = 12)			p-value ^a
	Frequency of detection	Mean	SD	Frequency of detection	Mean	SD	
Ametryn	15	0.75	1.09	12	0.19	0.28	0.038*
Atrazine	15	0.32	0.70	12	0.03	0.03	0.062
Pendimethalin	15	0.57	1.03	12	0.04	0.04	0.030*
2,4-D	5	0.004 ^b	0.004	4	0.003 ^b	0.001	–

^a p-values were calculated using a two-sample *t*-test.

^b For calculating averages, the proxy value (half of LOD) was assigned for samples $< \text{LOD}$.

* $p < 0.05$.

to and surrounded by sugarcane fields and our results suggest that the presence of pesticides in non-applicator households could be due to spray drift from sprayed nearby fields. Factors influencing spray drift and distance traveled include the application method and equipment, weather conditions, the amount of pesticide used, the distance from the source to the sampling site and topography (Weppner *et al.*, 2006; Coronado *et al.*, 2011a). Studies have shown that concentration of pesticide decreases with increasing distance from the spraying area (inverse correlation) (Simcox *et al.*, 1995; Kawahara *et al.*, 2005; Coronado *et al.*, 2011a; Brouwer *et al.*, 2018). Our results are consistent with previous investigations demonstrating that pesticide drift can reach households within 400 m from the spraying area (Fenske *et al.*, 2002; Koch *et al.*, 2002; Siebers *et al.*, 2003; Quandt *et al.*, 2004; Carlsen *et al.*, 2006) and another study that detected pesticide residues 750 m from the spraying area (Ward *et al.*, 2006).

Curwin *et al.* (2005) reported that atrazine and 2,4-D were below detectable levels in all air samples collected from farm and non-farm homes, in Iowa, USA. This could be possibly because some homes in that study were located at around 800 m from the sprayed fields and thus the concentrations were very low at the sampling sites (homes). Moreover, the air samples were collected 7 days (first visit) and 30 days (second visit) after the application of pesticides in the field. In contrast, in our study, households were much closer to the sprayed fields (~200 m) and we collected indoor air samples on the day of pesticide application. It would therefore be

reasonable to expect that our data would show higher levels of pesticides. In another study, mean pesticide concentrations ranged from $< \text{LOD}$ to $0.023 \mu\text{g m}^{-3}$ in homes of non-applicators in an agricultural community in Japan (Kawahara *et al.*, 2005). These are much lower than our results, as our mean concentration for all four target compounds from non-applicator households ranged from $< \text{LOD}$ to $0.19 \mu\text{g m}^{-3}$ (Table 2). The lower pesticide levels in the Japanese investigation can be explained by the fact that some of the homes were situated more than 400 m from the sprayed field. A USA-based study involving a farm housing community in central Washington investigated the role of off-target pesticide drift as a pathway for human exposure in nearby communities (Weppner *et al.*, 2006). The households in that study were closer to the nearest sprayed fields than the households in our study, which is reflected by the much higher reported pesticide levels in outdoor air from the non-applicator households in the USA study (median: $0.48 \mu\text{g m}^{-3}$) than our indoor air results ($< \text{LOD}$ – $0.19 \mu\text{g m}^{-3}$). Moreover, households in the USA study were surrounded by potato, corn and wheat fields and were within 15–200 m of the nearest treated field, while the highest concentration ($0.98 \mu\text{g m}^{-3}$) was recorded from a household located within 10–20 m from the nearest field. Study evidence also shows that the likelihood of spray drift increases during the summer, as a result of spray volatilization due to high temperatures (Kawahara *et al.*, 2005; Gibbs *et al.*, 2017). Sprayed pesticides evaporate at high temperatures during summer months and partially attach to suspended air particles, then diffuse and transfer through air to reach off-target sites. Our study samples were collected during summer, when household windows and doors were open during the day because of high temperatures (mean afternoon temperatures: 28.6°C and 31.1°C) while pesticide spraying and sample collections were conducted (Table S1), so pesticides sprayed in the fields potentially penetrated indoors. Wind conditions were calm during the pesticide applications, although we did not measure wind speed due to lack of instrumentation, as detailed in our previous paper (Msibi *et al.*, 2021); elevated temperatures were a contributing factor to household exposure through spray drift.

Table 2 presents a comparison of published data from studies performed in countries outside Africa that have analyzed pesticide concentrations in indoor and outdoor air in agricultural communities. Compared with the published data, the mean concentrations from our study are higher than those in studies from Japan (Kawahara *et al.*, 2005) and the USA (Whitmore *et al.*, 1994; Lu *et al.*, 2004; Curwin *et al.*, 2005; Gibbs *et al.*, 2017). This is mainly due to the fact that in comparison with most of the studies in Table 2, our sampling sites (households) were much closer to the sprayed fields, as we noted earlier. However, one study from the USA reported higher indoor air concentrations in non-applicator households. Whitmore *et al.* (1994) investigated indoor air

Table 2. A comparison of indoor pesticide concentrations in this study with those in published data from other countries.

Country	Target compound	Mean concentrations ($\mu\text{g m}^{-3}$)		Sampling location	Sample size	References
		Applicator household	Non-applicator household			
Eswatini	4 pesticides	$< \text{LOD}$ –0.75	$< \text{LOD}$ –0.19	Indoor air	27 households	This study
Japan	7 pesticides	–	$< \text{LOD}$ –0.023	Indoor and outdoor air	55 households	Kawahara <i>et al.</i> (2005)
Iowa, USA	6 pesticides	–	$< \text{LOD}$ –0.05	Indoor air	20 homes	Curwin <i>et al.</i> (2005)
Washington, USA	2 pesticides and their oxygen analogs	3×10^{-5} – 5.4×10^{-3}	$< \text{LOD}$ – 2×10^{-4}	Indoor and outdoor air	13 households	Gibbs <i>et al.</i> (2017)
Washington, USA	Methamidophos	–	0.05–0.48	Indoor and outdoor air	6 households	Weppner <i>et al.</i> (2006)
Florida, USA; Massachusetts, USA	22 pesticides	–	$< \text{LOD}$ –0.529	Indoor air	–	Whitmore <i>et al.</i> (1994)
Washington, USA	5 pesticides	2×10^{-3} – 4×10^{-3}	–	Indoor air	6 homes	Lu <i>et al.</i> (2004)

pesticide levels in non-applicator households at two study sites in the USA. Their indoor air concentrations range from $< \text{LOD}$ to $0.529 \mu\text{g m}^{-3}$ and are higher than our findings from non-applicator households, although it is worth noting that the households in that USA study also reported the use of pesticides on indoor plants and the use of insecticides to control household pests (Whitmore *et al.*, 1994). Gibbs *et al.* (2017) investigated indoor and outdoor air concentrations of chlorpyrifos and azinphos-methyl, with their analogs, in households in an agricultural region of central Washington, USA. They found higher indoor air levels of the target pesticides in farmworker households with close proximity to tree fruit fields (≤ 250 m from the nearest tree fruit field) compared with non-proximal households (> 250 m from the nearest tree fruit field). In that study, the reported indoor air concentrations for proximal households ranged from 3×10^{-5} to $5.4 \times 10^{-3} \mu\text{g m}^{-3}$, much lower than our findings ($< \text{LOD}$ – $0.19 \mu\text{g m}^{-3}$). In our study, we collected air samples on the day of pesticide application in the fields, whereas the Washington study could not confirm that the target pesticides had been sprayed in the nearby tree fruit field during the sampling period (Gibbs *et al.*, 2017).

3.1.2 Take-home pathway

As there was little variance in household proximity to the sprayed fields in our study, we assert that the significantly higher pesticide levels in applicator versus non-applicator households can be ascribed to applicators bringing the pesticide residues home with them on the clothes that they had worn during spraying activities. On the days of indoor air sample collection, all target pesticides had been sprayed in the morning by applicators who participated in our previous investigation into applicators' pesticide inhalation exposures. After spraying activities were concluded for the day, the applicators were followed to their homes for the collection of indoor air samples. The high concentrations of pesticide residues detected in their households provide evidence of potential secondary exposure to pesticides among family members. Notably, pesticide levels were significantly higher in applicator households than in non-applicator households for ametryn (0.75 vs. $0.19 \mu\text{g m}^{-3}$) and pendimethalin (0.57 vs. $0.04 \mu\text{g m}^{-3}$) (Table 1). For atrazine, the difference in concentrations between applicator and non-applicator households was marginally significant. At 25°C , the vapor pressures of our target compounds are 9.40×10^{-6} mmHg for pendimethalin, 2.74×10^{-6} mmHg for ametryn, 2.89×10^{-7} mmHg for atrazine and 1.39×10^{-7} mmHg for 2,4-D (HSDB, 2020). Thus, it would be expected that the highly volatile pesticides (ametryn and pendimethalin) would easily vaporize and travel through air onto the skin, clothes and shoes of the applicators, and also enter the nearby households via drift conditions. This would explain why our results show that ametryn and pendimethalin have the highest levels detected in both sets of households. It was difficult to estimate the amount of each pesticide sprayed in the fields on each spraying day. Moreover, it is likely that unequal amounts of each pesticide were sprayed in the sugarcane fields. Thus, the different airborne concentrations of the pesticides detected in our samples reflect the amounts of pesticides sprayed. The results in this paper are consistent with the levels of pesticide residues in applicators' personal air samples in our previous paper, which reported the highest detection rates and mean concentrations for ametryn and pendimethalin and the lowest detection rate and concentrations for 2,4-D amongst all four pesticides studied (Msibi *et al.*, 2021). Since 2,4-D was only detected in 33.3% of each type of household, 2,4-D was not detected frequently enough ($> 50\%$) to allow for meaningful analysis of statistical significance of the difference between households. Our data in Table 1 suggest that the take-home pathway is the most influential source of indoor agricultural pesticides and demonstrate much higher indoor concentrations of ametryn and pendimethalin in applicator households compared with those in non-applicator households.

Our observations are consistent with previous studies that illustrate significant increases in residential exposure to agricultural pesticides via the take-home pathway (Curl *et al.*, 2002; Hogenkamp *et al.*, 2004; Coronado *et al.*, 2006; Thompson *et al.*, 2014; Tamaro *et al.*, 2018). For example, Golla *et al.* (2012) found that workers on farms using a commercial pesticide applicator to spray atrazine had lower levels of the pesticide inside their homes than farmers who applied the pesticide themselves. In a study from Iowa in the USA, concentrations of several pesticides were more than 4-fold higher in homes of agricultural workers compared with homes without agricultural workers (Ward *et al.*, 2006). Other researchers have reported finding pesticides solely

used in agriculture, such as atrazine and metolachlor, only in households of applicators spraying these particular pesticides (Curwin *et al.*, 2005; Arcury *et al.*, 2007; Hyland and Laribi, 2017). In addition, biomonitoring studies have shown that children in households of pesticide applicators exhibit statistically higher concentrations of metabolites than children from households without farm workers (Fenske *et al.*, 2000; Lu *et al.*, 2000; Lambert *et al.*, 2005; Coronado *et al.*, 2006). In a study that investigated concentrations of organophosphate (OP) pesticide residue in house dust from an American agricultural community, Lu *et al.* (2000) reported that levels of the pesticide metabolites in the urine of farmworkers' children were significantly higher than levels in the urine of non-farmworkers' children. They also found a marginally significant association between house dust concentrations and urinary metabolite concentrations of OP pesticides in farmworker families (Lu *et al.*, 2000). In another USA study, Lozier *et al.* (2012) reported lower levels of atrazine inside homes where farmworkers removed their shoes before entering the home compared with homes where shoes were worn inside. Conversely, other studies have shown no evidence of pesticide exposure via the take-home pathway (Thompson *et al.*, 2008; Coronado *et al.*, 2012; Salvatore *et al.*, 2015; González-Alzaga *et al.*, 2018).

Most studies investigating exposure to agricultural pesticides via the take-home pathway have broadly focused on farmworker households. For example, one study (Lu *et al.*, 2004) conducted in Washington, USA, focused on farmworker households, whilst our study specifically focused on applicator households. The study by Lu *et al.* (2004) investigated the presence of five OP pesticides in agricultural homes, which were defined as having at least one adult who worked in an agriculture-related job. The study reported mean indoor air concentrations ranging from 2×10^{-3} to $4 \times 10^{-3} \mu\text{g m}^{-3}$ (Lu *et al.*, 2004), which are much lower in comparison with our findings, in which indoor concentrations from applicator households ranged from $< \text{LOD}$ to $0.75 \mu\text{g m}^{-3}$. In addition, they found pesticide residues in house dust and children's hands and toys in agricultural homes, providing more evidence of household contamination via the take-home pathway (Lu *et al.*, 2004). Pesticide applicators using spraying equipment in the fields are more exposed to pesticides than other farmworkers such as harvesters and machine operators; therefore, it would be expected that applicators would transfer higher levels of pesticide residues into their homes.

3.2 Health Risk Assessment

After completing the probabilistic risk assessment, we obtained HQ_i distributions representing the non-cancer risk due to inhalation exposures for each of the four pesticides in all age groups (Table 3). In a comparison of both sets of data, household residents were most likely to be exposed to ametryn and least of all to 2,4-D. The risk of exposure was up to 4 times higher in applicator households than in non-applicator households. Children aged 1–3 years were consistently at the highest risk of exposure to all target compounds, with mean HQ values for ametryn, atrazine, pendimethalin and 2,4-D in applicator households of 0.358, 0.033, 0.053 and 5.4×10^{-4} , respectively. In applicator households, HQ values at the 95th percentile (HQ₉₅) for children aged 0.5–0.9 and 1–3 years exceeded 1 for ametryn (i.e., 1.001 and 1.101, respectively) (see Table 3 and Fig. S2). This finding indicates a significant health risk resulting from ametryn inhalation exposure among children in applicator households. However, this risk may be compounded by various other factors, including exposures through dermal absorption and ingestion of pesticide residues in the environment. Coscolla *et al.* (2017) investigated pesticide levels in outdoor ambient air in a rural French community and reported a maximum HQ value for pendimethalin of 9.29×10^{-5} for infants (6 months–1.5 years). In comparison, our data revealed an HQ₉₅ value of 0.01 for the risk of exposure to pendimethalin among infants aged 0.5–0.9 years in non-applicator households. Recent studies describing risk assessments of inhalation exposure to pesticides have reported higher HQ values for infants and children compared with values among adults (Li *et al.*, 2014; Yusà *et al.*, 2014; Coscolla *et al.*, 2017). Notwithstanding, all of those reported HQ values were less than 1, which is considered an acceptable risk.

At the 95th percentile, the cumulative risk of exposure to all target compounds was > 1 in applicator households for children aged 0–3 years, which is concerning, as such a risk is considered unacceptable (Fig. 3). In contrast, in the non-applicator households, the mean and 95th percentile cumulative risk was acceptable at < 1 for all age groups. It is worth noting that our results represent the high-end risk of exposure; families in the applicator households may not be

Table 3. HQ values due to indoor exposures to the target compounds.

Age group (years)	Inhalation rate (m ³ day ⁻¹)	Body weight (kg)	Applicator households			Non-applicator households		
			Mean	95 th percentile	SD	Mean	95 th percentile	SD
<i>Ametryn</i>								
< 0.5	4.1	7.4	0.308	0.944	0.290	0.083	0.252	0.078
0.5–0.9	5.4	9.2	0.325	1.001	0.308	0.087	0.267	0.082
1–3	8.9	13.8	0.358	1.101	0.338	0.096	0.294	0.090
4– 2	12	31.8	0.209	0.642	0.198	0.056	0.172	0.053
13–18	15.2	56.8	0.149	0.456	0.140	0.040	0.122	0.037
19–65	16.3	68.1	0.133	0.409	0.125	0.036	0.108	0.033
> 65	14.2	68.1	0.116	0.355	0.109	0.031	0.095	0.030
<i>Atrazine</i>								
< 0.5	4.1	7.4	0.029	0.111	0.040	0.003	0.008	0.002
0.5–0.9	5.4	9.2	0.030	0.117	0.042	0.003	0.008	0.002
1–3	8.9	13.8	0.033	0.128	0.046	0.003	0.009	0.003
4–12	12	31.8	0.020	0.075	0.027	0.002	0.005	0.002
13–18	15.2	56.8	0.014	0.053	0.019	0.001	0.004	0.001
19–65	16.3	68.1	0.012	0.047	0.017	0.001	0.003	0.001
> 65	14.2	68.1	0.011	0.041	0.015	0.001	0.003	0.001
<i>Pendimethalin</i>								
< 0.5	4.1	7.4	0.045	0.160	0.053	0.003	0.008	0.002
0.5–0.9	5.4	9.2	0.048	0.170	0.056	0.003	0.008	0.002
1–3	8.9	13.8	0.053	0.186	0.061	0.003	0.009	0.003
4–12	12	31.8	0.031	0.109	0.036	0.002	0.005	0.002
13–18	15.2	56.8	0.022	0.077	0.025	0.001	0.004	0.001
19–65	16.3	68.1	0.020	0.069	0.023	0.001	0.003	0.001
> 65	14.2	68.1	0.017	0.060	0.020	0.001	0.003	0.001
<i>2,4-D</i>								
< 0.5	4.1	7.4	5.0×10^{-4}	0.002	6.2×10^{-4}	3.9×10^{-4}	0.001	3.7×10^{-4}
0.5–0.9	5.4	9.2	5.2×10^{-4}	0.002	6.6×10^{-4}	4.1×10^{-4}	0.001	4.0×10^{-4}
1–3	8.9	13.8	5.8×10^{-4}	0.002	7.3×10^{-4}	4.6×10^{-4}	0.001	4.4×10^{-4}
4–12	12	31.8	3.4×10^{-4}	0.001	4.2×10^{-4}	2.7×10^{-4}	0.001	2.6×10^{-4}
13–18	15.2	56.8	2.4×10^{-4}	0.001	3.0×10^{-4}	1.9×10^{-4}	0.001	1.8×10^{-4}
19–65	16.3	68.1	2.1×10^{-4}	0.001	2.7×10^{-4}	1.7×10^{-4}	0.001	1.6×10^{-4}
> 65	14.2	68.1	1.9×10^{-4}	0.001	2.3×10^{-4}	1.5×10^{-4}	0.001	1.4×10^{-4}

exposed to the high levels of the target pesticides reported in this study every day throughout the year. However, we highly recommend that after completing spraying activities, applicators should change and wash their personal protective equipment (PPE), shower, and wash all clothing worn while spraying, before they leave the workstation. Such measures will minimize the risk to households posed by take-home pesticides.

3.3 Limitations of the Study

This study describes finding high levels of agricultural pesticides in households on the days of spraying in nearby farms; the main contributors to these high levels were take-home and spray drift pathways. We only assessed residential inhalation exposure to four pesticides, so did not account for other routes of exposure, such as dermal and ingestion routes, despite being aware that the household members are also exposed to pesticides through dust, water, soil and food. Quantifying these multiple co-exposures would more comprehensively cover levels of non-occupational exposure in the households. In addition, our analysis may have underestimated the cumulative risk of inhalation exposure to pesticides, as residents are exposed to other pesticides used in the study area. Studies investigating the presence of pesticides in house dust have detected pesticide residues in carpet dust and on children's hands and their toys (Lu *et al.*, 2000; Curl *et al.*, 2002; Lu *et al.*, 2004). Young children are inclined to touch the floor and other surfaces

Fig. 3. HI values at 95th percentile showing the cumulative risk for each age group for all four target compounds. * Hazard index > 1.

as they crawl and play with their toys, so are very likely to ingest dust through hand-to-mouth contact (Lu *et al.*, 2004). As our study did not investigate pesticide levels in house dust, we may have underestimated the level of exposure and the resulting cumulative health risk. Longitudinal monitoring of the study area would provide greater understanding of the complexity and changes in pesticide levels through different seasons and pathways. Furthermore, biomonitoring data would have been useful, as biomarkers represent the summation of exposure and could provide an estimate of the internal doses for family members and the associated adverse health effects.

4 CONCLUSIONS AND RECOMMENDATIONS

Our findings, which detail the risks posed by indoor residential inhalation exposure to pesticides in Swazi agricultural communities, may serve as a basis for future regulatory and behavioral interventions in African countries. We measured significantly higher levels of agricultural pesticides in the applicators' homes than in the non-applicators' homes, with young children in applicators' households being the most vulnerable to exposure. As the distances from all of the residences to the adjacent sugarcane fields were similar, the concentrations we found may have been influenced by pesticide residues transported home by the applicators. We therefore urge farms and pesticide applicators to implement more stringent measures for mitigating exposure via the take-home pathway. Our results and recommendations may be valid for other African nations where farmworker families typically reside near agricultural land. Comprehensively assessing this population's household exposure to pesticides, i.e., from all environmental media, warrants further research. Moreover, epidemiological studies must be conducted in such communities.

DISCLAIMER

The authors declare no conflicts of interest.

SUPPLEMENTARY MATERIAL

Supplementary material for this article can be found in the online version at <https://doi.org/10.4209/aaqr.210062>

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