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Exposure risk to rural Residents: Insights into particulate and gas phase pesticides in the Indoor-Outdoor nexus

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ABSTRACT

Rural residents are exposed to both particulate and gaseous pesticides in the indoor-outdoor nexus in their daily routine. However, previous personal exposure assessment mostly focuses on single aspects of the exposure, such as indoor or gaseous exposure, leading to severe cognition bias to evaluate the exposure risks. In this study, residential dust and silicone wristbands (including stationary and personal wearing ones) were used to screen pesticides in different phases and unfold the hidden characteristics of personal exposure via indoor-outdoor nexus in intensive agricultural area. Mento-Carlo Simulation was performed to assess the probabilistic exposure risk by transforming adsorbed pesticides from wristbands into air concentration, which explores a new approach to integrate particulate (dust) and gaseous (silicone wristbands) pesticide exposures in indoor and outdoor environment. The results showed that particulate pesticides were more concentrated in indoor, whereas significantly higher concentrations were detected in stationary outdoor wristbands ($p < 0.05$). Carbendazim and chlorpyrifos were the most frequently detected pesticides in dust and stationary wristbands. Higher pesticide concentration was found in personal wristbands worn by farmers, with the maximum value of 2048 ng g^{-1} for difenoconazole. Based on the probabilistic risk assessment, around 7.1 % of farmers and 2.6 % of bystanders in local populations were potentially suffering from chronic health issues. One third of pesticide exposures originated mainly from occupational sources while the rest derived from remoting dissipation. Unexpectedly, 43 % of bystanders suffered the same levels of exposure as farmers under the co-existence of occupational and non-occupational exposures. Differed compositions of pesticides were found between environmental samples and personal pesticide exposure patterns, highlighting the need for holistic personal exposure measurements.

1. Introduction

In order to meet the global food demand of a growing population, farmers use pesticides to increase crop production, using nearly 2.7 million tons of active substances in 2020 (FAO, 2022). Despite its vital contribution to securing food availability (Tang et al., 2021), the massive input of pesticides has caused ubiquitous contamination in environmental matrices in fields and residential areas (Geissen et al., 2021; W. Jiang et al., 2016; Jiang & Gan, 2012; Weiying Jiang et al., 2016; Mu et al., 2023). Pesticide exposure may result in accumulation in body tissues and contribute to multiple health problems, including cancer, asthma, diabetes, Alzheimer's disease and reproductive issues (Huang et al., 2019; K.M. Hayden, 2010; Kumar, 2004; Lina S. Balluz, 2000; Rusiecki et al., 2006; Velmurugan et al., 2017). Approximately 44 % of global farmers are facing the consequences of pesticide exposure which is responsible for a human death rate between 0.4 and 1.9 % (Boedeker et al., 2020; Hassaan & El Nemr, 2020). Thus, concerns have been raised regarding the potential pesticide exposure risk for rural residents.

Rural residents can be exposed to both particulate and gaseous pesticides present in their surroundings. Specifically, pesticide exposure may occur via inhalation of gaseous pesticides in the ambient air, ingestion of pesticide-contaminated dust or direct skin contact with pesticide drifts or particles (Koelmel et al., 2022; Mu, Zhang, et al.,

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2022). Individual exposure risks from indoor and outdoor environments can differ due to the different degradation rates of pesticides which can be caused by several factors including wind, humidity, temperature, and solar radiation (W. Jiang et al., 2016). Unfortunately, most studies related to pesticide risk exposure assessments focus mainly only on individual aspects of exposure, such as indoor or outdoor exposure (Degrendele et al., 2022; Msibi et al., 2021; Mu, Zhang, et al., 2022; Waheed et al., 2017), which may lead to substantial discrepancies when compared with actual combined exposures.

Given the highly individualized daily routines of farm workers and the differing agricultural tasks they carry out, measuring direct personal pesticide exposure is challenging. An active air sampling technique using a pump and collection device which actively absorbs ambient air is often used to measure personal exposure to airborne chemicals, especially in occupational settings (Estill et al., 2020; Nguyen et al., 2022; NIOSH, 2018). However, the sampler is burdensome to use and thus may interfere with accurate participant measurements. Biomonitoring examines internal exposure to certain chemicals by analyzing biological samples, such as hair, urine, milk, and blood plasma, but this sampling procedure can be invasive (Henríquez-Hernández et al., 2022; Huber et al., 2022; LaKind et al., 2009; Melissa Legrand, 2005; Thompson et al., 2023). Silicone wristbands, with the ability to absorb volatile and semi-volatile chemicals, have been used as low-cost samplers for a wide range of airborne contaminants, reflecting personal exposure profiles primarily from inhalation and partial dermal contact (deposition of contaminated dust and drifts on skin) (Hendryx et al., 2020; Kile et al., 2016; Nguyen et al., 2022; O'Connell et al., 2014; Samon et al., 2022). Due to their ease of use, silicone wristbands have been given to residents (Aerts et al., 2017), children (Kile et al., 2016; Kim G Harley, 2019; Koelmel et al., 2022), industrial workers (Hendryx et al., 2020; Nguyen et al., 2022), and even pets (Catherine F Wise, 2020; Wise et al., 2021) to monitor their exposure to hundreds of chemicals including pesticides, polycyclic aromatic hydrocarbons, flame retardants and polychlorinated biphenyl. O'Connell built predictive models for silicone wristbands to translate the measured concentrations in wristbands to the equivalent air concentrations under equilibrium conditions, which provides a new approach to assess gas phase contaminants (O'Connell et al., 2021). By transforming pesticide measurements into air concentrations, wristbands can serve as wearable sensors to address the spatial variances of pesticide levels across the indoor-outdoor nexus and to obtain real individual exposure to gaseous pesticides during daily routines. Currently, exposure assessment studies rely mostly on active/passive sampling programs that neglect exposure resulting from highly individualized daily routines (Liu et al., 2022; Mamontova & Mamontov, 2022). Consequently, there is an urgent need to integrate the personal exposure profiles collected from silicone wristbands with the results of risk assessments.

The North China Plain is a major grain producing area in China, accounting for only 3 % of the total national land area but contributing to one third of the total national pesticide input (NBS, 2020). The intense pesticide use in this region has led to soil contamination and potential risks to ecological endpoints (Mu, Wang, et al., 2022; Mu et al., 2023). Pesticides were frequently misused in the study region with excessive amounts of fungicides application observed (Mu, Wang, et al., 2022). Meanwhile, multiple residues were detected in surface soils, moreover, nearly half of monitored sites showed high ecological risks to soil biota (Mu et al., 2023). To date, little is known about the exposure risk of pesticides to rural residents in this region. To address this, we performed a probabilistic exposure risk assessment of pesticides for the local populations in this region, integrating major exposure routes using a Mento-Carlo simulation. The objectives of this study were: 1) to investigate the occurrence of particulate and gas phase pesticides in the indoor-outdoor nexus via dust and wristbands measurements; 2) to obtain the individualized pesticide exposure profiles using wristbands; and 3) to assess the probabilistic health risks of pesticides for the local population by integrating the major exposure routes to particulate and gaseous pesticides in the indoor-outdoor nexus. This study expanded on former exposure assessments by using direct measurements collected from personal wristbands and comparing these measurements to the equivalent air concentrations and inhalation risks, thereby determining comprehensive pesticide exposure risk for local residents.

2. Materials and methods

2.1. Study design

Prior to sampling, farmer interviews were carried out in Quzhou, a typical agricultural county in the North China Plain (NCP), to investigate the usage patterns of pesticides in different farming systems. During the interviews, farmers who plant apples and grapes are prone to using more pesticides, as well as reporting more frequently misuse behaviours (Mu, Wang, et al., 2022). To explore more details of exposure risks of pesticide, those group of farmers were selected for the wristband experiment. To make sure the selected farmers could be representative, some extra criteria were concerned: 1) participants should be willing to attend the wristband experiment and follow the sampling protocols, 2) participants must be scattered in villages nearby the main streets, and 3) participants should be registered in the villages and they are continuously living in the village, thereby having a regular daily routine within or around the villages. In total, 35 participants were recruited, including 21 farmers and 14 bystanders. Forty-six (46 %) of participants were female, aged from 29 to 66. Among them, thirty-eight (38 %) of recruited farmers were reported to have applied pesticides more than twice during the monitoring period. When preparing and using pesticides, farmers were told to use self-protective measures, such as gloves and water-proof cloths, to avoid direct contact with pesticide drifts. Based on the interview results, 24 commonly used pesticides were selected and analyzed in this study. Usage patterns of selected pesticides were listed in the supplementary materials (Table S1). The names, chemical groups, molecular mass, and Chemical Abstracts Service (CAS) numbers are summarized in Table S2.

Along with the wristbands participants were given to wear around their wrists, additional wristbands were placed in their homes (hanging in the living room) and in outdoor environments close to fields (only for farmers) to monitor the background pesticide exposure levels in domestic and field environments, respectively. Given that pesticides can keep low diffusion rates in silicone wristbands in summer for a month with promising recovery rates (Anderson et al., 2017), the monitoring period for wristbands was set at 4 weeks to monitor personal exposure during the peak summer season for agricultural activities. Participants were informed that they could take off their wristbands during showers and when sleeping, and they were assured that they could quit the experiment at any time. During the first and last days of the experiment, dust samples were collected from the floors with a vacuum cleaner (T10 mix, Puppy Electronic Appliances Internet Technology Beijing Co., Ltd.) in indoor (living room) and outdoor (main street or pavement surface near houses) locations for each participant. After sampling, dust and any other garbage collected by the vacuum were transferred to a prepared self-sealing bag and transported to the lab. In the lab, each collected sample was first passed through a 0.15 mm sieve to separate surface dust from larger particles. All wristbands and sieved dust samples were placed in self-sealing bags and stored at −20℃ until analysis could be completed.

2.2. Pesticide determination

2.2.1. Pre-treatment, chemicals, and solvents

Collected dust samples were passed through a 0.15 mm sieve to remove other materials such as hair, stones and tiny pieces of domestic garbage and then placed into self-sealing bags stored at −20℃. Standard adult size wristbands (20 cm L × 1.2 cm W × 0.2 cm T) were purchased online (https://www.1688.com/). Before use, a precleaning procedure was conducted to eliminate possible interference from unexpected chemicals (O'Connell et al., 2014). The wristbands were cleaned in a shaker followed by a two-step procedure: 1) 30 min extraction using ethyl acetate and a hexane solvent (1:1, v: v) and 2) 30 min extraction using ethyl acetate and a methanol solvent (1:1, v: v). After the extraction, wristbands were dried under a nitrogen stream and placed in the freezer (4℃).

The analytical reference standards of analyzed pesticides were purchased from Alta Scientific Co., Ltd. For the determination of dust samples, the standard stock solution and mixed standard solution were prepared in acetonitrile at concentrations of 1000 and 100 mg/L. The calibration curve for instrumental analysis was prepared by diluting the mixed standard solution to reach the concentrations of 0.01, 0.05, 0.1, 0.5, 1 and 2 mg/L in acetonitrile. For the determination of wristband samples, the mixed standard solution was prepared at a concentration of 0.1 mg/L. A standard ^{13}C-caffeine solution was used as an internal standard. The calibration curve for instrumental analysis was prepared by diluting the mixed standard solution to reach the concentrations of 1, 5, 20, 50 and 100 μg mL^{-1} in acetonitrile. All solutions were stored in the refrigerator at −20℃ until use.

2.2.2. *Pesticide extraction and instrumental analysis*

The analytical method for dust samples was modified and based on the QuEChERS (Quick, Easy, Cheap, Effective, Rugged and Safe) method (Michelangelo Anastassiades, 2003). Briefly, dust samples, along with water, acetonitrile and NaCl, were vortexed and centrifuged for pesticide extraction. $MgSO_4$ and C_{18} were then added into centrifuge tubes with extracts to remove interfering substances. The upper layer supernatants were filtered and transferred into glass vials.

Wristband samples were spiked with ethyl acetate and ^{13}C caffeine. The samples were then mixed and transferred to evaporation flasks and evaporated until dryness using dimethyl sulfoxide (DMSO) as a keeper. The remaining solvents were reconstituted and filtered into LC vials by adding acetonitrile and ultrapure water. All samples were stored at –20℃ while awaiting instrumental analysis. Full details of the pretreatment of dust and wristband samples are provided in Text S1 in the supplementary materials.

Instrumental measurements were carried out using liquid chromatography coupled with LC-MS/MS with a triple quadrupole mass spectrometer (Shimadzu LCMS-8045, Shimadzu Corporation, Tokyo, Japan) and a Shimadzu LC system coupled to a triple quadrupole mass spectrometer QTRAP (6500+, Sciex, Canada) for dust samples and wristbands, respectively. Full details of instrumental analysis procedures for dust and wristband samples are provided in Text S2, Table S3 and Table S4 in the supplementary materials.

2.2.3. *Quality assurance and quality control*

To avoid possible cross contamination during the dust sampling process, the electric motor of the vacuum cleaner was removed, and the remaining parts of the vacuum cleaner were thoroughly washed by hand with soap and water between each sampling interval. Prior to the lab analysis, untreated bare soil samples were collected in Quzhou and passed through a 0.15 mm sieve to serve as blank samples. The blanks were then fortified with the mixed standard solution at concentrations of 0.01, 0.05, 0.1, 0.5 and 1 mg/L for recovery assessment and method validation.

For wristband samples, additional worn wristbands were precleaned and used as blank samples. The blanks were then fortified with the mixed standard solution at concentrations of 0, 0.2, 0.5, 1, 5 and 10 μg L^{-1} for recovery assessment and method validation. The calibration curve solutions were injected after 10 successive sample injections to recalibrate the machine. Recovery efficiencies of analyzed pesticides were acquired within a range of 70 % to 110 %, except for thiophanate-methyl which was excluded from the measurement list. The calibration curves obtained good linearity with the correlation coefficients over 0.99.

The calibration curve solutions were injected at the beginning of the measurements and again after each 10 successive sample injections. Deviations of the analytical results of each calibration curve solution sample were within 30 %. Recovery efficiencies of analyzed pesticides for fortified blank samples and calibration curve solutions were both acquired within a range of 70 % to 110 %. The calibration curves obtained good linearity with the correlation coefficients over 0.99.

2.3. *Pesticide risk assessment*

In this study, the chronic lifetime exposure risk of pesticides for individuals was assessed based on the health risk assessment method. In the assessment, Hazard Quotients (HQs) were calculated for individual exposure routes and further summed up as a Hazard Index (Feng et al.). For a HI > 1, the daily exposure could result in chronic health risks, otherwise the risk can be considered negligible. The assessment was conducted mainly based on the measurements taken from dust samples and personal wristbands. Specifically, measurements of indoor and outdoor dust samples were run through the model to assess the exposure risk from dust / particle ingestion and dermal contact routes in the indoor and outdoor environments. Wristband results mainly represented pesticide inhalation risks of participants as they carried out their daily routines. It should be noted that pesticide concentrations in the wristbands represent only the pesticides taken up by the silicone within a fixed period. In this case, concentrations collected from personal wristbands were converted into air concentrations for the sake of the exposure assessment.

2.3.1. *Equivalent air concentrations*

The amount of pesticides absorbed by the wristbands increased over time before reaching a constant exposure level, following a dose–response relationship (Bartkow et al., 2005). The uptake process of chemicals by passive samplers, such as wristbands, can normally be divided into three phases, including kinetic (linear), intermediate (curvilinear) and equilibrium (Feng et al., 2022). To calculate equivalent concentrations for certain passive samplers under equilibrium conditions, well-established quantitative models are available which are based on Fick's first law of diffusion (O'Connell et al., 2021). This model adapts to the uptake of chemicals from any phase and follows rate constant-based equations (Equation (1) and (2).

$$C_a = \frac{N_{compound}}{V_s \times K_{sa} \times \left(1 - e^{(-k_e \times t)}\right)} \tag{1}$$

or

$$C_a = \frac{N_{compound}}{V_s \times K_{sa} \times \left(1 - e^{-\frac{R_S \times t}{V_S \times K_{sa}}}\right)} \tag{2}$$

where C_a represents the equivalent air concentration of pesticides converted from wristband concentrations. $N_{compound}$ and Vs represent the amount (mass) of pesticides in the sampler and the volume of the sampler, respectively. The wristbands used in this study were the regular adult sized version, which was approximately 5.30 g and 0.00445 L of silicone (O'Connell et al., 2021). R_s and k_e are rate-based parameters that stand for the sampling and dissipation rate, respectively. K_{sa} is a partitioning coefficient demonstrating the ratios between concentrations in the passive sampler and ambient environment matrix at equilibrium during the deployment stage.

Boiling point (BP)-based models, including the BP-TEST (toxicity estimation software tool, Equation (3) and (4) and the BP-OPERA (open structure–activity/property relationship app, Equation (4) and (5) models were considered as the best performance model and secondary model, respectively. The predicted BPs and other calculated model parameters are listed in Table S5.

$$logk_e = -0.012 \times BP(^{\circ}C - TEST) + 2.04 \tag{3}$$

$$logK_{sa} = 0.02BP(^{\circ}C - TEST) + 0.517 \quad (4)$$

$$logk_{e} = -0.009 \times BP(^{\circ}C - OPERA) + 1.55 \quad (5)$$

$$logK_{sa} = 0.019 \times (BP^{\circ}C - OPERA) + 0.9 \quad (6)$$

2.3.2. *Probabilistic health risk assessment*

The chronic exposure risks of pesticides are assessed based on the health risk assessment model developed by the US Environmental Protection Agency (USESA). This model requires inputs of the environmental concentrations of chemicals and exposure parameters, which are normally set as the mean or maximum values of corresponding parameters to give deterministic assessment results. The deterministic assessments from former studies present general-case or worst-case scenarios for the exposure risk, potentially causing elevated uncertainties and leading to under- or overestimates of the health risk. Thus, this study takes the uncertainties of each input variable into account and provides probabilistic risk assessments of the local populations by using a Mento-Carlo simulation.

The processing procedure of the Mento-Carlo simulation mainly includes: (1) setting random variables and inputting the corresponding distribution; (2) setting simulation variables; and (3) running the model for 10,000 iterations at a 95 % confidence level (Yuan et al., 2023). The probabilistic health risks were calculated for farmers and bystanders separately. To present the best estimates of the underlying risks, pesticide exposure was separated into 5 different sections for each group of the population: daily inhalation (personal wristband data), indoor ingestion, indoor dermal exposure, outdoor ingestion, and outdoor dermal exposure. Since individuals spend over 80 % of their daily lives in an indoor environment, a time-weighted exposure frequency was set to determine the best estimates of the exposure risks under simulations of realistic exposure scenarios. Lists of random variables and their distributions, including pesticide concentrations and exposure parameters, are summarized in the supplementary information from Table S6 to Table S8.

2.4. *Statistical analysis*

Descriptive statistics for all samples were conducted using SPSS (version 26; IBM, USA). The Kolmogorov-Smirnov test was used for the normality test. The one-way ANOVA and Mann-Whitney U tests were used to compare means between the pesticide concentrations of samples from different subgroups. Origin 2021 was used to draw the box plots and the lollipop chart. The heatmap of personal pesticide exposure profiles was created using R programming language (lattice package). For the multivariant analysis, partial least squares-discriminant analysis (PLS-DA) was conducted using MetaboAnalyst (NSERC, 2022) to visualize and compare pesticide exposure between farmers and bystanders. Raw data was transformed to mean centered and lognormal prior to the analysis. The ellipses in the biplot represent the 95 % confidence level of the two groups of participants. Variable importance in projection (VIP) scores, quantifying the impacts of each of the predictor variables to the response variable, were calculated to determine the contributions of the components to the distinctive exposure characteristics of farmers and bystanders. The Mento-Carlo simulation was performed to assess the probabilistic health risks of pesticides for the local populations using the Oracle Crystal Ball version 11.

3. Results and discussion

In this study, we monitored pesticide concentrations in dust samples and stationary wristbands to determine the personal pesticide exposure of rural residents in the indoor-outdoor nexus. Over the course of 4-weeks, we collected 136 dust samples and retrieved 77 % of the distributed wristbands (n = 70) from farmers and bystanders as well as some that were hanging in indoor and outdoor locations.

3.1. *Occurrence of pesticides in particulate and gas phases in the indoor-outdoor nexus*

3.1.1. *Levels of pesticides in dust*

Multiple pesticide residues were found in all dust samples (Fig. S1A) with 3 to 24 residues detected in each sample. Notably, over 40 % of samples contained more than 15 pesticide residues. Measured concentrations were widely distributed in 4 orders of magnitude in dust with the highest concentrations reaching 50345 ng g^{-1} (for atrazine from an indoor location). Carbendazim, thiamethoxam and acetamiprid were the most frequently detected pesticides and measured in more than 90 % of the samples. The presence of pesticides in dust differed largely among the different locations. More residues and higher concentrations of residues were found in indoor dust samples (Fig. 1A, B). There were on average 17 and 13 pesticide residues detected in indoor and outdoor dust samples, respectively. The accumulation levels of pesticides in indoor dust were 1.3 to 25 times higher than those of outdoor dust, except for atrazine, chlorantraniliprole and lufenuron (Table 1). Pesticides were more frequently detected and found in higher concentrations in the dust samples from locations surrounding farmers' residences (Fig. 1C, D). The geomeans of pesticide concentrations were between 1.02 and 20 times higher in the samples from farmers than those from bystanders, except for the concentrations of atrazine, carbendazim, tebuconazole and prochloraz.

This study revealed the distribution pattern of pesticides in indoor and outdoor dust collected from farmers and bystanders. Pesticides in residential dust mainly originated from the wind facilitated transport of pesticide contaminated soil particles from adjacent fields, and the take-home pathway (Dereumeaux et al., 2020). In particular, the take-home pathway by farmers has been found to be a nonnegligible contributor to their non-occupational exposure via transferring pesticides from contaminated clothes, shoes, and skin to the residential environments (López et al., 2019). Despite not handling pesticides on their own, bystanders have more complex daily routine and higher possibilities for non-occupational pesticide exposures compared to farmers. Thus, pesticides in residential dust collected from bystanders may also originate from the take-home pathway, especially for those who incidentally had contact with pesticides in their daily routine. In this study, higher concentrations of carbendazim, atrazine, and prochloraz were found in dust samples collected at locations around bystanders (Fig. 1C). These pesticides were commonly used in the greenbelt along the road near the village, which might be exposed to bystanders and further transferred into residential area through the take-home pathway. The unexpected distribution pattern of certain pesticides between farmers and bystanders revealed that all residents living in the intensive farming regions are potentially suffering from pesticide exposure.

Despite the variances in the sampling locations and seasons, these results were consistent with those of previous studies performed in other regions but had much higher outliers (S. Mukerjee, 1997; Simaremare et al., 2021; Velázquez-Gómez et al., 2019) (Table S9). As one of the most detected pesticides, chlorpyrifos was measured at much higher concentrations in the indoor dust with a maximum value at 15463 ng g^{-1}, which is 1.6 and 141 times higher than studies carried out in Taiwan province and California, USA (Barbara J Mahler, 2009; Hung et al., 2018), respectively.

3.1.2. *Levels of pesticides in stationary wristbands*

Pesticides were detected in all stationary wristbands with the number of residues ranging from 2 to 19. Up to 65 % and 33 % of samples were determined to contain more than 5 and 10 pesticide residues, respectively (Fig. S1B). Atrazine and chlorpyrifos were the most frequently detected pesticides showing up in more than 90 % of samples. The largest concentration of a pesticide found in a stationary wristband sample was that of tebuconazole which was detected at 6475 ng g^{-1} from an outdoor location. As compared to dust samples, wristbands from outdoor locations exhibited significantly higher pesticide levels (Fig. 1A,

Table 1
Measured concentrations (ng/g) of pesticides in dust and wristbands in the indoor and outdoor environments.

Pesticides	Indoor dust		Outdoor dust		Indoor wristbands		Outdoor wristbands	
	Geomean (Detection rate, %)	Range	Geomean (Detection rate, %)	Range	Geomean (Detection rate, %)	Range	Geomean (Detection rate, %)	Range
Abamectin	227.700 (71.0)	ND-2152.30	42.7000 (26.9)	ND-215.000	5.00000 (96.3)	ND-58.4000	5.20000 (30.8)	ND-17.2000
Acetamiprid	1570.80 (97.1)	ND-293122	113.500 (88.1)	ND-9854.20	3.20000 (3.7)	ND-3.20000	16.1000 (92.3)	ND-1601.00
Atrazine	126.000 (75.4)	ND-50345.6	171.400 (53.7)	ND-26122.5	3.20000 (33.3)	ND-27.9000	15.2000 (100)	ND-92.5000
Carbendazim	1579.90 (98.6)	ND-61928.0	469.300 (100)	17,0—22305.3	10.0000 (18.5)	ND-27.3000	29.4000 (76.9)	ND-231.100
Carbofuran	80.4000 (63.8)	ND-1127.30	52.3000 (74.6)	ND-182.900	2.80000 (48.1)	ND-20.5000	8.00000 (100)	ND-217.800
Carbofuran 3Hydroxy	40.1000 (17.4)	ND-426.400	14.3000 (3.0)	ND-17.0000	NA	NA	2.00000 (15.4)	ND-3.40000
Chlorantraniliprole	76.7000 (56.5)	ND-566,800	180.100 (10.4)	ND-1299.00	NA	NA	3.90000 (15.4)	ND-4.30000
Chlorobenzuron	856.900 (94.2)	ND-44850,6	260.900 (83.6)	ND-12268.9	8.60000 (40.7)	ND-202,300	31.0000 (100)	ND-1701.40
Chlorpyrifos	400.100 (97.1)	ND-15463,9	96.8000 (89.6)	ND-807.700	7.10000 (96.3)	ND-246,2	41.2000 (100)	ND-989.800
Clothianidin	94.7000 (91.3)	ND-2707,40	30.0000 (16.4)	ND-273.200	1.70000 (3.7)	ND-1,7	1.30000 (7.7)	ND-1,30000
Difenoconazole	2099.50 (24.6)	ND-25204,2	102.300 (91.0)	ND-26500.0	3.10000 (22.2)	ND-21,6	10.7000 (92.3)	ND-404.400
Dimethomorph	453.400 (85.5)	ND-29336,0	71.2000 (88.1)	ND-2900.00	6.70000 (29.6)	ND-29,3	97.3000 (76.9)	ND-1156.00
Fipronil	97.8000 (33.3)	ND-2020,30	39.1000 (4.5)	ND-200.000	2.90000 (14.8)	ND-12,2	10.6000 (69.2)	ND-175.200
Fipronil sulfone	72.3000 (44.9)	ND-824,400	34.6000 (10.4)	ND-63.0000	2.30000 (3.7)	ND-2,30000	4.50000 (69.2)	ND-69.2000
Imidacloprid	2044.00 (98.6)	ND-71390,3	101.100 (91.0)	ND-8255.40	2.60000 (33.3)	ND-11,9000	9.50000 (84.6)	ND-345.700
Lufenuron	43.2000 (75.4)	ND-638.600	157.100 (1.5)	ND-157.100	6.20000 (3.7)	ND-6.20000	5.20000 (7.7)	ND-5.20000
Nicosulfuron	75.2000 (92.8)	ND-12401.3	23.2000 (61.3)	ND-274.300	NA	NA	NA	NA
Pendimethalin	80.9000 (73.9)	ND-2134.70	50.5000 (89.6)	ND-159.200	2.00000 (48.1)	ND-5.90000	9.60000 (100)	ND-533.200
Prochloraz	182.200 (60.9)	ND-17268.2	74.2000 (35.8)	ND-6800.00	5.00000 (3.7)	ND-5.00000	3.50000 (7.7)	ND-3.50000
Propamocarb	185.000 (10.1)	ND-4372.80	159.900 (22.4)	ND-6527.00	1.20000 (3.7)	ND-1.20000	2.20000 (7.7)	ND-2.20000
Pymetrozine	153.500 (53.6)	ND-46502.7	119.300 (10.4)	ND-4474.80	NA	NA	NA	NA
Pyridaben	152.700 (42.0)	ND-30794.0	118.500 (22.4)	ND-598.600	NA	NA	3.70000 (15.4)	ND-7.70000
Tebuconazole	1689.90 (47.8)	ND-20513.5	142.400 (67.2)	ND-9124.80	7.70000 (44.4)	ND-123.300	63.8000 (100)	ND-6475.00
Thiamethoxam	859.500 (91.3)	ND-859943	87.8000 (94.0)	ND-6268.10	5.80000 (18.5)	ND-18.7000	3.40000 (38.5)	ND-6.10000

Note: ND, not detected; NA, not applicable.

B). Higher concentrations of all pesticides were detected in the stationary wristbands from outdoor locations and the geomeans for individual pesticides were 1.04 to 14.5 times higher than those from indoor wristbands (Table 1 and Fig. 1D).

This study found much higher pesticide levels in stationary wristbands located in outdoor environments, which could be attributed to the successive outdoor pesticide applications occurring during the monitoring period. Similar trends were observed by Aerts where more pesticides were detected in the wristbands from outdoor locations rather than indoor locations (Aerts et al., 2017). There were no significant differences in the pesticide concentrations between stationary wristbands collected from farmers and bystanders through Mann-Whitney *U* test (Fig. 1D), indicating that bystanders may have suffered the same level of inhalation risk from pesticides inside their homes, despite the fact they had not had any direct exposure to pesticide use for a long period.

3.1.3. Comparisons between pesticides detected in dust and wristbands

More residues and higher concentrations of residues were measured in dust samples as compared to stationary wristbands from both indoor and outdoor environments (Fig. 1A, B). The mean concentrations and number of residues from dust samples (2325 $\mu g\ kg^{-1}$ and 15 residues) were 12.5 and 1.5 times higher than those from wristbands, respectively. Pesticides were found in all samples. Over 90 % of dust samples contained more than 10 residues, while only 37.5 % of wristband samples had such levels. For individual pesticides, measured concentrations were found to differ across locations and mediums (Figure. S2). Specifically, 7 out of the 24 pesticides found in dust, such as abamectin, imidacloprid and chlorpyrifos, varied significantly across the indoor-outdoor nexus and were found in significantly higher concentrations in indoor locations with at least 2-fold differences. For pesticides found in wristbands, higher concentrations of atrazine, acetamiprid, tebuconazole, chlorpyrifos and pendimethalin were found in outdoor stationary wristbands. The varied pesticide distribution patterns among collected samples indicates that particulate pesticides tended to

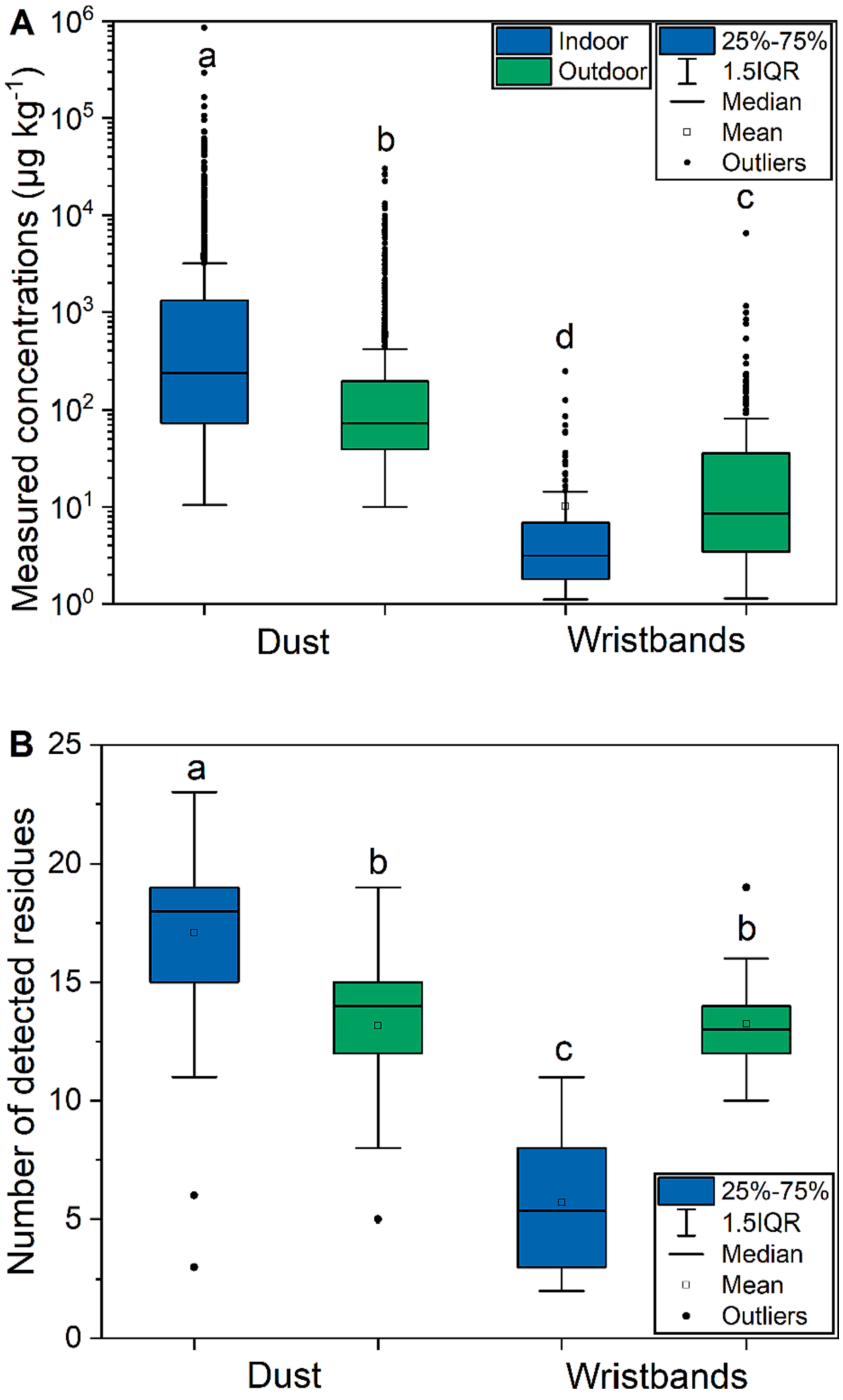

Fig. 1. Pesticide levels found in dust and wristbands in the indoor and outdoor nexus: A, pesticide concentrations in dust and wristbands collected from indoor and outdoor locations; B, number of detected residues in dust and wristbands from different locations; C, lollipop chart of the geomeans of pesticide concentrations in indoor and outdoor dust; D, lollipop chart of the geomeans of pesticide concentrations from indoor and outdoor stationary wristbands. Figures in red font in A and B represent the overall means and medians of dust and wristband samples. IQR, interquartile range. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

accumulate in indoor environments, whereas gaseous pesticides accumulated more in outdoor spaces. It should be noted that pesticides commonly used or reported being used at excessive dosage in this area, were all found to be heavily accumulated in both dust and stationary wristband samples. Imidacloprid and acetamiprid, the most frequently used pesticides with usage frequencies exceeding 40 %, were shown up in more than 90 % and 50 % of dust and stationary wristband samples, respectively. Farmers grow vegetables and apple orchards applied

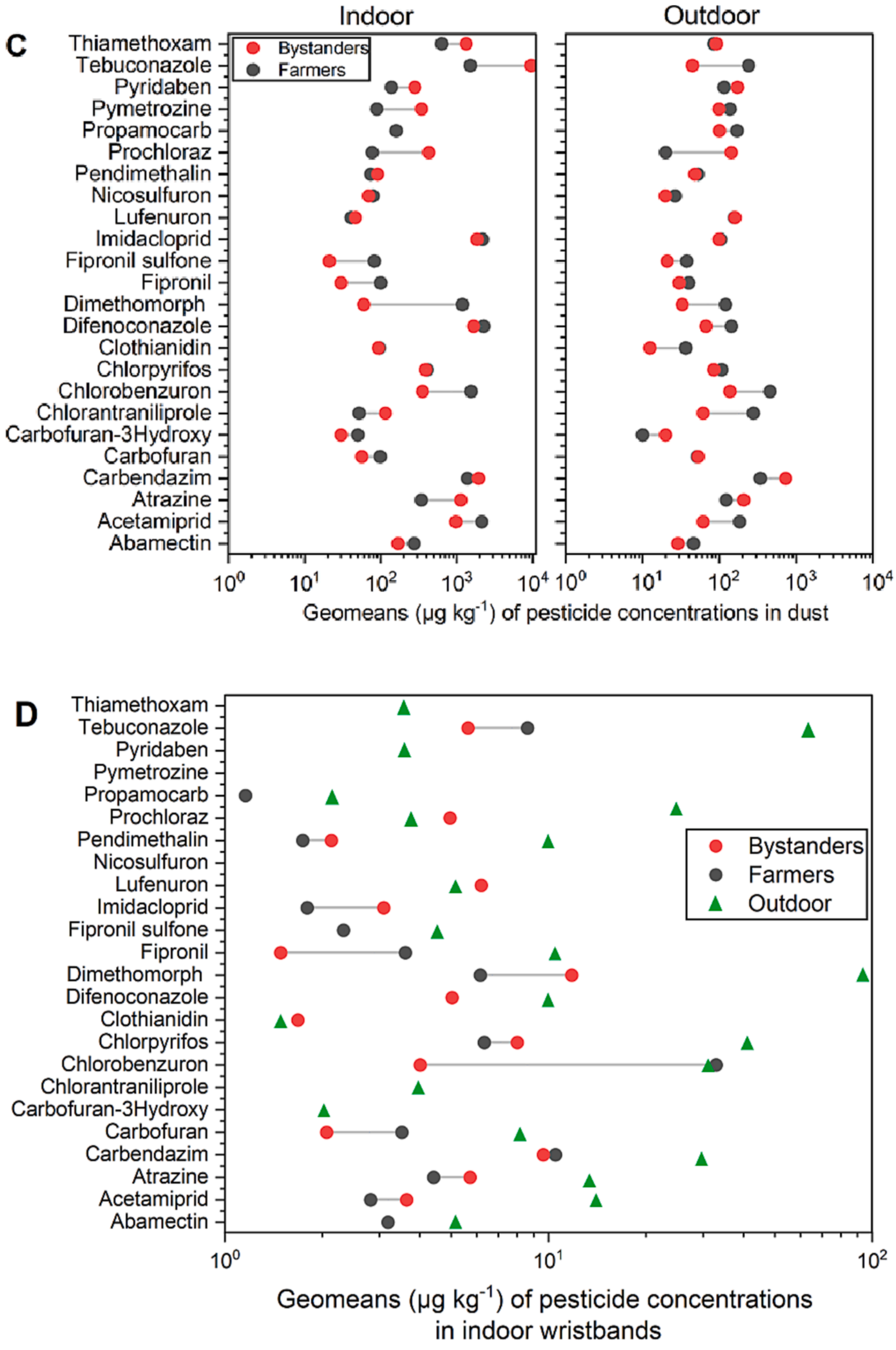

Fig. 1. (*continued*).

substantial amounts of dimethomorph and difenoconazole to prevent plant diseases, leading to much higher concentrations at monitored sites.

The cumulative patterns of pesticides found in the different locations varied depending on what phase the pesticide was detected in (Figure. S3). Despite low usage frequency (Fig. 2A), chlorpyrifos was the most abundant pesticide found in both dust and wristband samples, with detection frequencies exceeding 90 % in the two mediums. This was followed by atrazine. Chlorpyrifos, targeting primarily the central and peripheral nervous systems, is recognized as a class II moderately hazardous pesticide by the World Health Organization (WHO, 2020). Along with having negative effects on the nervous system, exposure to chlorpyrifos has been associated with acute poisoning effects such as eye irritation, dermatological defects, endocrine disruption and cardiovascular disease (David L Eaton, 2008; Ubaid ur Rahman et al., 2021). Similarly, atrazine was found to cause toxic effects on the nervous system by inducing cerebellar toxicity (Chevrier et al., 2011). In addition, particular attention should be given to prenatal exposure to atrazine, which was found to cause adverse birth outcomes (Xia et al., 2017). The widespread existence of these compounds raises concerns for the environmental exposure risks from pesticides in our surroundings and highlights the need for a shift in pesticide use patterns in the major farming systems in the NCP.

3.2. *Personal pesticide exposure profiles*

The personal pesticide exposure from daily routines was monitored using silicone wristbands for 4 weeks during the peak summer season of pesticide applications. The number of detected residues in wristbands

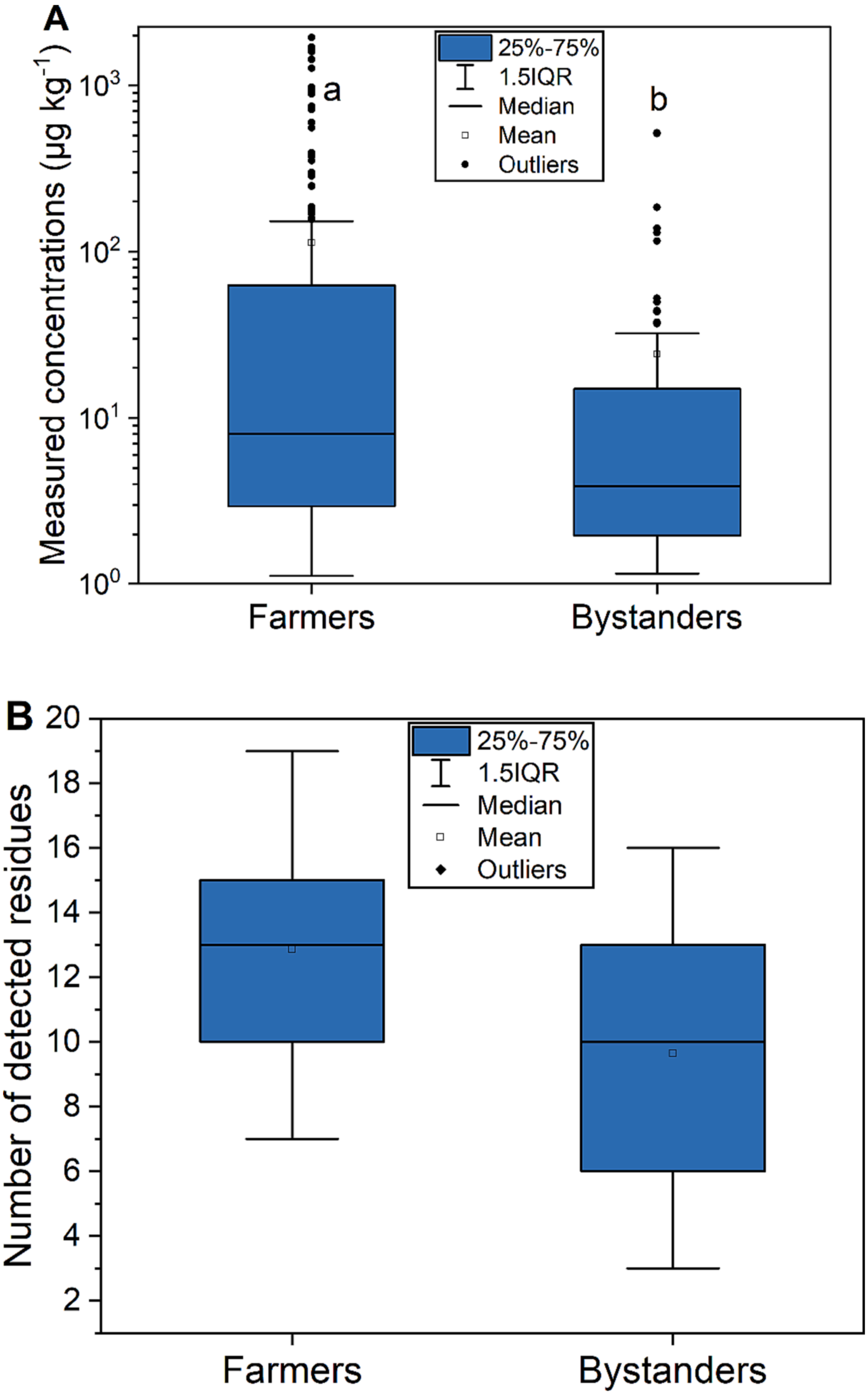

Fig. 2. Personal pesticide exposure profiles of farmers and bystanders: A, boxplot of measured pesticide concentrations in wristbands worn by farmers and bystanders; B, boxplot of number of detected residues in wristbands worn by farmers and bystanders; C, heatmap of measured pesticide concentrations in personal wristbands; D, biplot of the PLS-DA (partial least square discrimination analysis), ellipses represent the 95% confidence levels of two groups of participants; E, underlying exposure sources for individual pesticides based on VIP (variable importance in projection) scores. IQR, interquartile range.

worn by farmers (mean value of 13) was higher than that of bystanders (mean value of 10), yet the difference was not significant (Fig. 2A). More than 10 residues were detected in 88 % and 37 % of wristbands worn by farmers and bystanders, respectively. Significantly higher concentrations of pesticides were measured in wristbands worn by farmers as compared to those from bystanders (Fig. 2B, C). The maximum concentration of a pesticide found in personal wristbands was determined for difenoconazole at over 2000 ng g^{-1}, followed by dimethomorph and tebuconazole. The mean values of measured concentrations in wristbands worn by farmers were 1 to 17 times higher than those of bystanders, except for thiamethoxam and prochloraz (Table 2). It should be noted that both farmers and bystanders tended to be exposed to

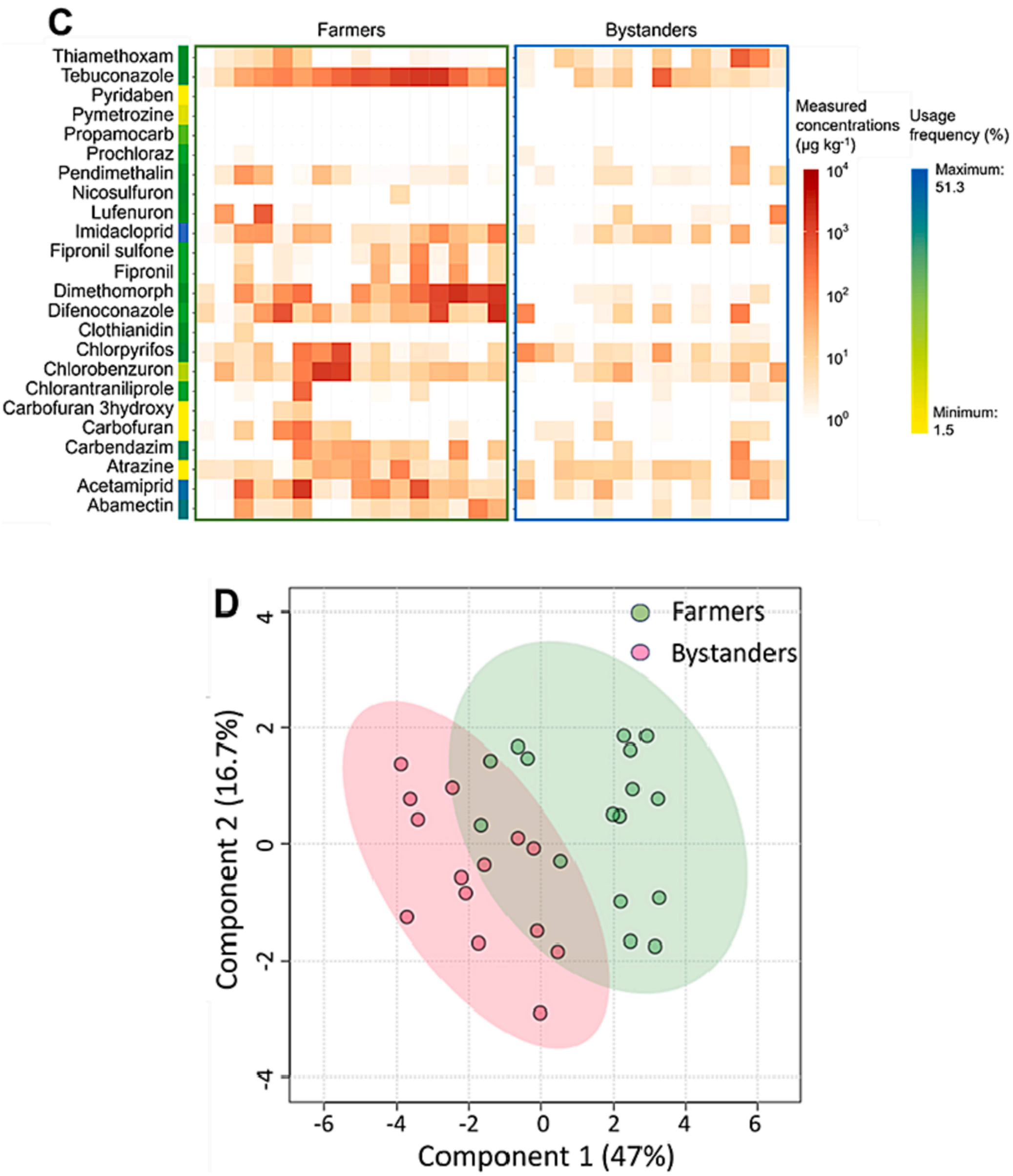

Fig. 2. (*continued*).

pesticides that were frequently used in the region at higher concentrations (Fig. 2C).

PLS-DA was used to study the differences between the personal pesticide exposure profiles of farmers and bystanders. The biplot allows us to see a clear distinction between the personal exposure profiles of farmers and bystanders (Fig. 2D). The slightly overlapping ellipses show different exposure patterns for most participants in the two groups. It is worth noting that the overlapping area between the ellipses indicates that 43 % of bystanders may have suffered the same level of pesticide exposure as farmers. VIP scores were further calculated to identify which pesticides contributed to the differences in the personal exposure patterns between farmers and bystanders (Fig. 2E). Difenoconazole and dimethomorph were evaluated as the pesticides that contributed the most to these differences with VIP scores exceeding 1.9. One-third of the analyzed pesticides showed VIP scores exceeding 1, indicating their significant contributions to the different exposure patterns.

The personal pesticide exposures of farmers and bystanders were characterized by a holistic silicone wristbands-dust approach. Particularly, pesticide concentrations in worn wristbands reflect the levels of airborne exposure in monitored periods. Despite protective measures taken by farmers during the pesticide preparation and application, the direct contact between pesticide drifts and wristbands cannot be totally avoided. But, practically speaking, to date, our approach could indicate the personal exposure risks of pesticide from airborne exposure. Farmers who applied pesticides more than 2 times suffered higher exposure risks than individuals came into fields less frequently with their average pesticide concentrations at 2373 and 722 ng g^{-1}, respectively. It is challenging to compare the pesticide concentrations due to lack of comparable studies with similar experiment settings, analytical list, and monitoring duration. Thus, the time-weighted concentrations were computed as ratios of the measured concentrations and the monitoring period (Table S10). For most of the compared pesticides (Aerts et al., 2017; Arcury, Chen, Quandt, et al., 2021; Fuhrimann et al., 2022; Kim G Harley, 2019), the current study detected higher pesticide concentrations, which is probably due to the longer monitoring period. Compared with former studies, participants in this study had higher daily exposure rates of fungicides including tebuconazole and carbendazim (Aerts et al., 2017; Fuhrimann et al., 2022) and lower exposure rates of chlorpyrifos (Arcury, Chen, Arnold, et al., 2021).

Farmers and bystanders had diverse exposure patterns, as exhibited

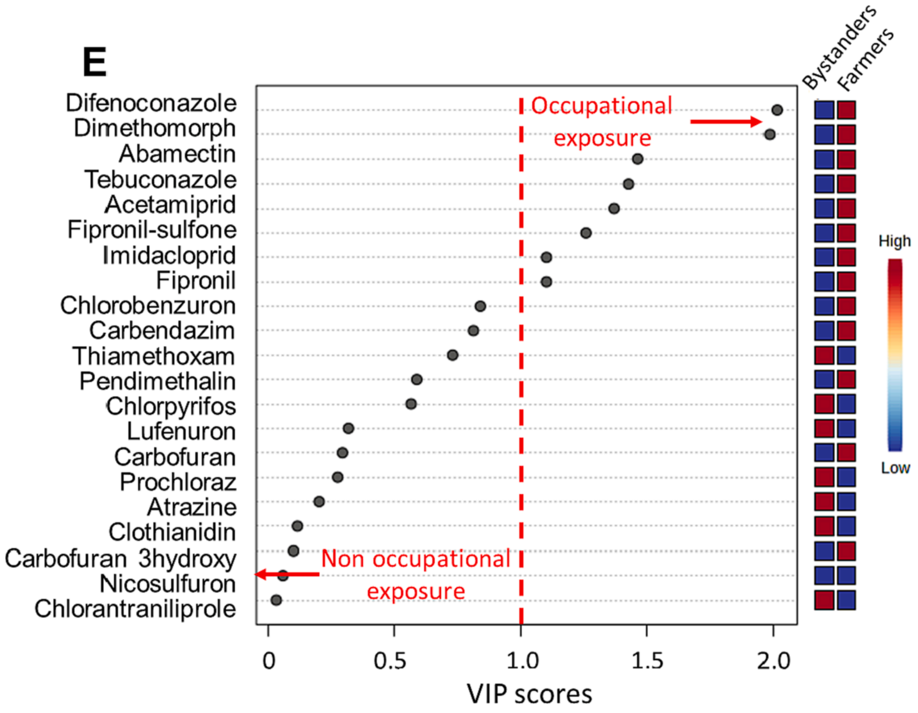

Fig. 2. (*continued*).

Table 2

Measured pesticide concentrations (ng/g) in wristbands worn by farmers and bystanders.

Pesticides	Overall			Farmers			Bystanders		
	Detection rate (%)	Geomeans	Range	Detection rate (%)	Geomeans	Range	Detection rate (%)	Geomeans	Range
Abamectine	53.30	7.400	ND-152.1	75.00	10.80	ND-152.1	28.60	2.400	ND-4.200
Acetamiprid	83.30	15.00	ND-1592	93.80	27.00	ND-1592	71.40	6.200	ND-49.70
Atrazine	90.00	10.00	ND-168.7	93.80	8.800	ND-168.7	85.70	11.70	ND-81.00
Carbendazim	40.00	25.50	ND-132.1	56.30	28.90	ND-132.1	21.40	17.60	ND-115.7
Carbofuran	43.30	7.600	ND-298.5	43.80	13.80	ND-298.5	42.90	3.800	ND-14.30
Carbofuran 3hydroxy	13.30	3.600	ND-11.60	12.50	8.200	ND-11.60	14.30	1.600	ND-2.300
Chlorantraniliprole	26.70	4.300	ND-409.8	25.00	7.600	ND-409.8	28.60	2.500	ND-3.900
Chlorobenzuron	90.00	12.80	ND-1590	93.80	24.60	ND-1590	85.70	5.600	ND-52.00
Chlorpyrifos	93.30	7.500	ND-888.6	87.50	9.400	ND-888.6	100	6.000	1,1–107.3
Clothianidin	13.30	2.400	ND-7.700	6.300	7.700	ND-7.700	21.40	1.600	ND-2.100
Difenoconazole	73.30	22.00	ND-2048	100.0	24.80	1,1–2048	42.90	15.90	ND-184.3
Dimethomorph	66.70	30.70	ND-1936	81.30	132.4	ND-1936	50.00	2.000	ND-4.200
Fipronil	30.00	8.300	ND-176.4	56.30	8.300	ND-176.4	0.000	NA	ND-NA
Fipronil sulfone	36.70	5.700	ND-126.6	68.80	5.700	ND-126.6	0.000	NA	NA
Imidacloprid	83.30	9.700	ND-185.4	93.80	13.20	ND-185.4	71.40	6.100	ND-44.20
Lufenuron	30.00	8.700	ND-554.4	18.80	38.70	ND-554.4	42.90	4.200	ND-126.8
Nicosulfuron	3.300	6.500	ND-6.500	6.300	6.500	ND-6.5	0.000	NA	NA
Pendimethalin	70.00	3.600	ND-79.90	81.30	3.500	ND-79.9	57.10	3.800	ND-29.30
Prochloraz	16.70	3.200	ND-37.90	12.50	1.300	ND-1.5	21.40	5.700	ND-37.90
Propamocarb	0.000	NA	NA	0.000	NA	NA	0.000	NA	NA
Pymetrozine	0.000	NA	NA	0.000	NA	NA	0.000	NA	NA
Pyridaben	0.000	NA	NA	0.000	NA	NA	0.000	NA	NA
Tebuconazole	90.00	49.50	ND-1707	100.0	151.6	1.4–1707.0	78.60	9.700	ND-515.8
Thiamethoxam	70.00	6.400	ND-511.6	62.50	3.700	ND-72.6	78.60	10.30	ND-511.6

Note: ND, not detected; NA, not applicable.

by the small overlapping area of the ellipses (Fig. 2D). The VIP scores (Fig. 2E) further showed that difenoconazole and other 7 pesticides with VIP scores exceeding 1 were drivers for the differing exposure patterns seen between farmers and bystanders. For exposure sources, personal pesticide exposure of farmers consists of occupational and non-occupational exposure, while the pesticide exposure of bystanders subjected to non-occupational sources. Thus, pesticides can be categorized into two groups: 1) 8 driver pesticides of the different exposure patterns between farmers and bystanders with VIP scores exceeding 1 that mainly originated from occupational exposure, and 2) other 16 pesticides with VIP scores lower than 1 that mainly came from non-occupational exposure. Furthermore, fungicides, including tebuconazole and dimethomorph, were the predominant pesticides that farmers were exposed to, whereas thiamethoxam contributed the most to the exposure of bystanders (Figure. S4). Given that these fungicides were used in significantly higher dosages in the study region (Mu, Wang, et al., 2022), they contributed substantially to the occupational exposure of farmers during pesticide applications. The overlap between the driver pesticides and the exposure contributors to farmers and bystanders indicates that the exposure to driver pesticides might have originated from occupational exposure, while the exposure to other pesticides could have originated from dissipated pesticide drifts from other regions. Despite the fact that bystanders did not participate in agricultural activities, the biplot of PLS-DA showed that approximately 43 % of bystanders had exposure patterns that were similar to farmers which means that they suffered the same level of health risks as farmers (Fig. 2D). In summary, for pesticide exposure, one-third can be attributed to occupational exposure and two-thirds can be attributed to diverse non-occupational sources. To some extent, the non-occupational exposure to a majority of the pesticides measured in this study poses the same health risks to farmers as to bystanders.

3.3. *Probabilistic health risk assessment of pesticides for residents*

This assessment addresses the following exposure concerns: (1) daily inhalation exposure; (2) indoor ingestion; (3) indoor dermal exposure; (4) outdoor ingestion and (5) outdoor dermal exposure. The ingestion and dermal exposures were assessed based on the occurrence of pesticides in indoor and outdoor dust. The assessment of daily inhalation exposures was carried out by translating measured concentrations from personal wristbands into the equivalent air concentrations under equilibrium conditions.

The equivalent air concentrations were calculated based on the BP-TEST and the BP-OPERA model (Table S11). Tebuconazole was the most abundant pesticide detected in daily routines with the highest concentration exceeding 1478 ng m^{-3}, followed by acetamiprid and chlorpyrifos. Furthermore, probabilistic health risks of pesticides for local populations were assessed using the Mento-Carlo simulation. The forecasts of the hazard index (Feng et al.) were computed for farmers and bystanders separately. Despite the fact that the 95 % CI of HIs for both farmers (from 2.8E to 03 to 9.6E-01) and bystanders (from 3.7E to 04 to 2.1E-01) were below the threshold levels, the probability of a possible chronic health risk to farmers and bystanders were approximately 7.1 % and 2.6 % (Figure. S5), respectively. As a high-density region of population, the NCP accounted for 3 % of the national land area with over 24 % of populations living in this region. Thus, the assessment results indicate that a large number of residents in this region are potentially suffering chronic exposure risk to pesticides even though relatively low proportions of farmers and bystanders were assessed at risk.

Sensitivity analysis was performed to identify critical factors affecting the forecasts for the HI (Figure. S6). For farmers, fipronil contributed the most to the sensitivity (over 60 %), followed by abamectin and chlorpyrifos, which in total accounted for over 82 % of the sensitivity. For bystanders, concentrations of carbofuran and tebuconazole contributed the most to the sensitivity (over 83 %). The probabilistic assessment revealed concerning levels of pesticide exposure risk to both farmers and bystanders. Compared to previous monitoring studies, the current study obtained higher HI forecasts (Hesami Arani et al., 2023; Vasseghian et al., 2022) and uncovered a potential chronic health risk for a small proportion of the general population by simulating realistic exposure scenarios and integrating all possible daily exposure routes.

3.4. *Implications and future study*

So far, this is the first study to characterize personal exposure to environmental pesticides covering major exposure routes including dermal, oral and inhalation. Consequently, there is a potential of chronic risk to populations living close to agricultural fields. The combined silicone-environmental medium approach could be a promising model to obtain individualized exposure profiles and determine personal exposure rates. Based on this workflow, epidemiological studies should be carried out to examine the links between pesticide exposure and health outcomes. Additionally, this study discovered highly distinct connections between personal and environmental exposures to pesticides in both dust and wristbands (Figure. S3 and S4). This finding reveals that the composition and abundance of pesticides in environmental samples taken from fixed sampling locations and within a fixed sampling radius probably cannot correctly mirror realistic personal exposure patterns, which highlights the need for more flexible and integrated personal exposure monitoring.

4. Conclusions

The present study revealed the distribution of pesticides in particulate and gaseous phases in the indoor-outdoor nexus sampling residential dust and silicone wristbands. The findings indicate that the daily pesticide exposure could pose chronic health risk to rural residents considering the ubiquitous of pesticides in surroundings, especially for farmers who are working with these compounds in farming practices. For the sources of pesticide exposure, only one third of the pesticides that participants exposed to originated from occupational path, the rest were from remote dissipation. Unexpectedly, around 43 % of the bystanders suffered the same level of pesticide exposure as the farmers due to the non-occupational exposure. This study explores a new approach to link personal exposure with environment quality which may help further study to understand personal exposure risk to environmental pollutants comprehensively.

CRediT authorship contribution statement

Hongyu Mu: Conceptualization, Methodology, Formal analysis, Visualization, Writing-Original draft. **Xiaomei Yang:** Project administration, Conceptualization, Supervision, Resources, Writing-Review & editing. **Kai Wang:** Supervision, Writing-Review & editing. **Rima Osman:** Methodology. **Wen Xu:** Resources. **Xuejun Liu:** Project administration, Supervision. **Coen J. Ritsema:** Supervision, Writing-Review & editing. **Violette Geissen:** Project administration, Resources, Supervision, Writing-Review & editing.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2024.108457.

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