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ORIGINAL PAPER

Risk assessment of airborne agricultural pesticide exposure in humans in rural China

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Abstract Continuous exposure to airborne pesticides causes their gradual accumulation in the human body, eventually posing a threat to human health. To the best of our knowledge, risk assessment study of pesticide non-occupational exposure to residents in agricultural areas has not been conducted in China. In this study, air samples (gas and dust) were collected from inside and outside residences of seven households and an area near the field in a grain-growing area (wheat and maize rotation) for eight months, and the pesticides present were examined both qualitatively and quantitatively. Using a 95% confidence interval, 9 out of 16 pesticides were detected, namely acetamiprid, acetochlor, atrazine, flucarbazone-sodium, imidacloprid, methyldisulfuron-methyl, nicosulfuron-methyl, pendimethalin, and beta-cyhalothrin, and their safety was subsequently evaluated. The results showed that the inhalation exposure of households to beta-cyhalothrin exceeded the acceptable range in the first residential, and the excess lifetime cancer risk of acetochlor inhalation exposure in six households and area around the field exceeds 1E−6, which highlights the need to strengthen preventive screening for cancer risk.

Keywords Pesticide · Inhalation exposure · Risk assessment · Environmental health outlook

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Introduction

Chemical pesticides are used globally as one of the most effective means of controlling diseases, insect pests, weeds, and rats (Sharma et al., 2020). Extensive use of chemical pesticides also poses risks to human health (Kirkhorn & Schenker, 2002, Stoytcheva, 2011; Freire et al., 2013; Siriporn et al., 2013; Luo et al., 2016; Brouwer et al., 2017), with infants and children being more susceptible to pesticides than adults (Koelmel et al., 2022). In the general application process, 50–70% of pesticides enter the air via volatilization and drift (Sui et al., 2010). Air is a good carrier of pesticide residues, and pesticides mainly exist in two forms, either as a gas or adsorbed by dust in the air. Pesticides in indoor dust remain stable over time owing to their protection from degradation by sunlight, fungi, and other factors. Pesticides in the air can enter the human body through the respiratory route, the dietary route, and the skin route (Lewis et al., 1994); therefore, it is crucial to assess air pesticide levels. Owing to pesticide-related air pollution can be readily ignored and the fact that people stay at home for a long part of the day, the possibility of pesticide exposure in indoor and outdoor environments is concerning (Pelletier et al., 2018).

Occupational and non-occupational exposure is the main modes of exposure to pesticides (Sabarwal et al., 2018). More attention should be paid to the long-term pesticide exposure caused by non-occupational exposure to chemicals present in the living environment. As the main component of non-occupational exposure, residential exposure is mainly caused by pesticide drift and use in both residences and farmlands (Nicole et al., 2015). There is a broader range of pesticides and higher concentrations present in agricultural residential areas compared with urban residential environments (Liaud et al., 2017); therefore, it is important to study the residential exposure of residents to pesticides in agricultural regions.

Recently published studies assess the risk of human exposure to air pollutants. A study conducted in Chile detected concentrations of polycyclic aromatic hydrocarbons in the air ranging from 2 to 108 ng/m^3 and assessed lifetime cancer risk (Pozo et al., 2023). Celine et al. assessed temporal and spatial variations in indoor dust in two agricultural areas of South Africa and detected more than 50 pesticides (Degrendele et al., 2022). A study of thiamethoxam inhalation exposure conducted in Seoul, South Korea, found that thiamethoxam entered the body through the respiratory route and was exposed to different concentrations in different parts (Lee et al., 2022). However, there have been no studies on pesticide inhalation exposure in China's vast agricultural areas, which is a worrying situation.

According to statistics from 2019, China consumes over 1,391,700 tons of pesticides, making it the nation with the highest pesticide use worldwide. In 2019, 120,300 tons of pesticides were used in Shandong Province in Eastern China, considered a typical agricultural region, which equates to 8.64% of the country as a whole (NBS, 2021). China has the largest agricultural population in the world; however, no research on the nation's pesticide inhalation exposure has been conducted. Therefore, we assessed the risk of daily pesticide non-occupational inhalation exposure in farmers living in the grain-growing regions of Shandong. We found a total of nine airborne pesticides and identified cancer/non-cancer-related risks in residents.

Materials and methods

Farmer selection and pesticide research

A representative grain crop-planting area in Nansun village (36°33′ 07″ N 116° 00′ 02″ E) in Shandong Province, China, was selected as the study site. Only wheat and maize are planted in this area. Seven typical households and an area near the field in the grain-growing area were selected to investigate pesticide used between March and October 2018. A total of 16 pesticides, all of which are currently used pesticides (CUP), were surveyed, and samples were collected monthly. The pesticides detected in this agricultural area were mesosulfuron-methyl, flucarbazone-Na, florasulam, imidacloprid, iminoctadine tris, MCPA-sodium, acetamiprid, abamectin, lambda-cyhalothrin, acetochlor, glufosinate-ammonium, nicosulfuron, atrazine, trifluralin, pendimethalin, and chlorpyrifos.

Sampling plan

In this study, we select passive air sampling devices that are more suitable for assessing the chronic toxicity of pesticides. It is mainly composed of polyurethane foam sampler (PUF-PAS) and metal shield, which can collect gas and dust in the air at the same time. It is almost impossible for human factors to affect this sampling process, which can meet the long-cycle sample collection work (Armstrong et al., 2014).

Factors affecting the non-occupational exposure to pesticides mainly include pesticide formulations, the distance between the residence and the field, the amount of pesticide applied around the residence, meteorological factors, hygienic factors, and the behavior of the occupants. We mainly considered the above factors when deploying samples and tried to avoid interference from factors unrelated to the experiment. PUF (140 mm × 13.5 mm) were deployed in the interior and exterior of seven residential buildings and in an area near farmland. To simulate human breathing, we placed the indoor sampler on a 1-m-high table, while the outdoor sampler was hung on a 1.5-m-high wall or wooden pole. Avoid contact with other objects as much as possible, and meteorological information was recorded simultaneously. Samplers were placed and collected on the 1st and 30th of each month. 159 PUF samples were collected over eight months, each sample contained three replications, dry ice is required for transportation. Samples are stored at minus 20 °C while being protected from light.

Sample analysis

The collected PUFs were then subjected to Soxhlet extraction. Determine the extraction solvent and extraction time in advance through pre-experiment. The solvent was ethyl acetate (100 ml) and the extraction time was 8 h. After Soxhlet extraction, the extract was concentrated to 1.5 ml using rotary evaporation, purified through an organic filter membrane, and then injected into a chromatographic sample vial. The qualitative analysis of the sample was carried out by GCMS-QP2020 (SHIMADZU) mass spectrometer, and the standard curve was made by the external standard method to quantify the sample. The feasibility of the experimental method was verified through addition and recovery experiments. The spike levels are 100 ppb, 1 ppm, and 5 ppm ($n=3$). The recovery rates for the three spiked levels are as follows: 86.69%, 81.67%, and 93.10%, and the relative standard deviation range was 1.57–7.32%. The column specification is Rtx-5MS (30 m × 0.25 mm × 0.25 μm). The chromatographic method is 50–160 °C, 30 °C/min; 160–200 °C, 15 °C/min; 200–270 °C, 10 °C/min; 270–300 °C, 5 °C/min. With splitless injection method, the injection port temperature is set to 250 °C and the injection volume is set to 2 μL.

Risk assessment calculations

The exposure concentration (EC) can be calculated using the following formula (Eq. 1):

$$EC = (CA \times ET \times EF \times ED)/AT \tag{1}$$

where EC (μg/m^3) is the exposure concentration; CA (μg/m^3) is the concentration of the pollutant in the air; ET (hours/day) is the exposure time; EF (days/year) is the exposure frequency; ED (years) is the exposure duration; AT [lifetime (years) × 365 days/year × 24 h/day] is the average time.

The HQ (hazard quotient) of the non-occupational inhalation route of pesticides is obtained by the following formula (Eq. 2):

$$HQ = EC/(\text{Toxicity value}^1 \times 1000\ \mu g/\text{mg}) \tag{2}$$

where HQ (no unit) is the danger quotient and toxicity values (mg/m^3) are inhalation toxicity values (e.g., RfC) appropriate for exposure scenarios. In this experiment, the toxicity value RfC (Reference Concentration) recommended by USEPA was selected.

To assess potential cancer risk, we primarily refer to the exposure factor manual (Agency, 2011) and the industry standard (ICAMA, 2017). As there are currently no pesticide-specific data on infants and children exposure susceptibility, we used ADAF (age-dependent adjustment factor) to estimate the additional cancer risk in early life exposure (EPA). The excess lifetime cancer risk can be calculated by the following formula (Eq. 3):

$$\begin{aligned} \text{Risk} = {} & (\text{IUR}_U\text{EC}_{<2} \times \text{ADAF}_{<2)}) \\ & + (\text{IUR} \times \text{EC}_{2-16} \times \text{ADAF}_{2-16}) \\ & + (\text{IUR} \times \text{EC}_{>16}) \end{aligned} \tag{3}$$

where EC (μg/m^3) is the exposure concentration and IUR (μg/m^3)$^{-1}$ is the unit risk of inhalation.

Results

Non-cancer-related risk assessment

Seven residences and an area around field in a rural community had potential non-cancer-related risks. A total of nine of the 16 pesticides previously surveyed were detectable: mesosulfuron-methyl, flucarbazone, imidacloprid, acetamiprid, lambda-cyhalothrin, acetochlor, nicosulfuron, atrazine, and pendimethalin. Choose the average CA (contaminant concentration in air) according to the actual data (Table 1). For each pesticide exposure scenario, based on the EPA and EU databases, the reported DAFs (dosimetric adjustment factors) were selected. Sensitive population factors and additional exposure risks for children and infants were also considered. The occupants spent 10 h indoors and 4 h outdoors, according to the data collected during preparation. The average EC values for the seven houses (L1–L7) and an area around fields (OF) in the agricultural region are shown in Fig. 1.

Table 1 Average CA of pesticides in grain-planting areas

Pesticide	L1 (μg/m^3)	L2 (μg/m^3)	L3 (μg/m^3)	L4 (μg/m^3)	L5 (μg/m^3)	L6 (μg/m^3)	L7 (μg/m^3)	OF (μg/m^3)
Acetamiprid	indoor4.66E−01 outdoor4.86E−01	indoor5.02E−01 outdoor4.82E−01	indoor4.50E−01 outdoor4.51E−01	indoor4.63E−01 outdoor3.48E−01	indoor4.18E−01 outdoor4.05E−01	6indoor 4.56E−01 6outdoor4.38E−01	7indoor4.65E−01 7outdoor4.18E−01	4.88E−01
Acetochlor	indoor1.14E+00 outdoor8.76E−01		indoor7.38E−02 outdoor3.60E−01	indoor5.11E−01	indoor9.61E−02 outdoor1.07E−01	indoor2.18E−01 outdoor5.23E−01		1.42E+00
Atrazine	indoor1.06E−03 outdoor3.28E−04	outdoor2.45E−04	indoor1.81E−04 outdoor1.57E−03	indoor9.33E−04 outdoor2.49E−03	indoor2.78E−03 outdoor1.40E−03	outdoor1.07E−03	indoor2.65E−03 outdoor1.65E−03	2.17E−03
Flucarbazone-sodium	indoor3.29E−02 outdoor3.30E−03	indoor6.62E−03 outdoor7.63E−03		indoor7.02E−03 outdoor3.49E−03	indoor4.97E−03 outdoor3.89E−03	indoor5.57E−04 outdoor1.58E−03	indoor1.86E−03 outdoor3.57E−02	1.28E−03
Cyhalothrin	indoor2.22E−01 outdoor4.98E−01		indoor1.16E−01 outdoor4.10E−01	indoor2.63E−01	indoor6.39E−02		indoor4.32E−01 outdoor1.98E−01	6.24E−02
Imidacloprid	indoor5.44E−02 outdoor3.90E−02	indoor9.42E−02 outdoor1.01E−01	indoor1.48E−01 outdoor8.24E−02	indoor1.13E−01 outdoor6.11E−02	indoor1.45E−01 outdoor1.21E−01	indoor1.05E−01 outdoor1.02E−01	indoor1.15E−01 outdoor7.12E−02	4.11E−02
Mesosulfuron-methyl	indoor1.25E−02 outdoor1.45E−02	indoor7.34E−03 outdoor2.51E−02	indoor1.48E−02 outdoor1.88E−02	indoor1.14E−02 outdoor7.38E−03	indoor1.32E−02 outdoor2.23E−02	indoor2.14E−02 outdoor1.54E−02	indoor1.48E−02 outdoor9.54E−03	1.17E−02
Nicosulfuron	indoor2.28E−02	indoor1.37E−01 outdoor7.36E−02	indoor5.83E−02 outdoor4.54E−02	indoor1.33E−02 outdoor1.10E−02	indoor2.64E−03 outdoor4.62E−02	indoor1.16E−01 outdoor3.45E−02	indoor1.52E−02 outdoor3.64E−02	3.46E−02
Pendimethalin	indoor1.06E−01	outdoor7.75E−02	indoor8.36E−02 outdoor7.72E−02	indoor1.25E−01 outdoor9.45E−02	outdoor8.24E−02	outdoor7.95E−02	outdoor8.93E−02	7.57E−02

L1—Resident 1; L2—Resident 2; L3—Resident 3; L4—Resident 4; L5—Resident 5; L6—Resident 6; L7—Resident 7; OF—Area near the field; a blank space indicates not detected

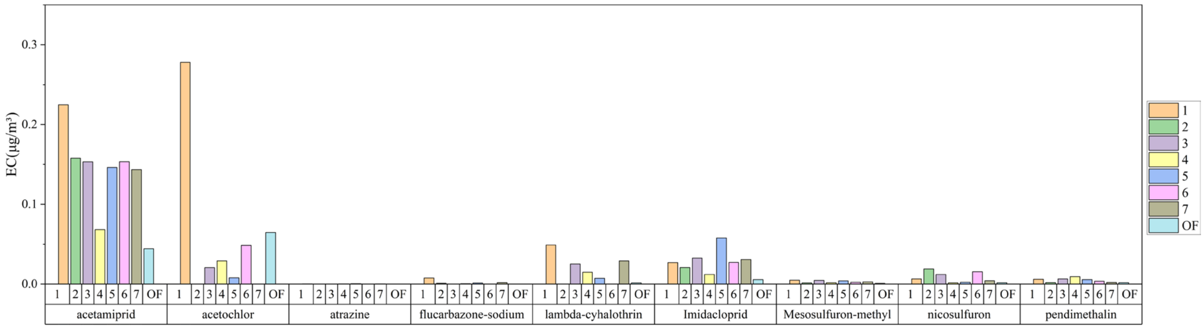

Fig. 1 Average exposure concentration of seven households and an area around fields in agricultural regions

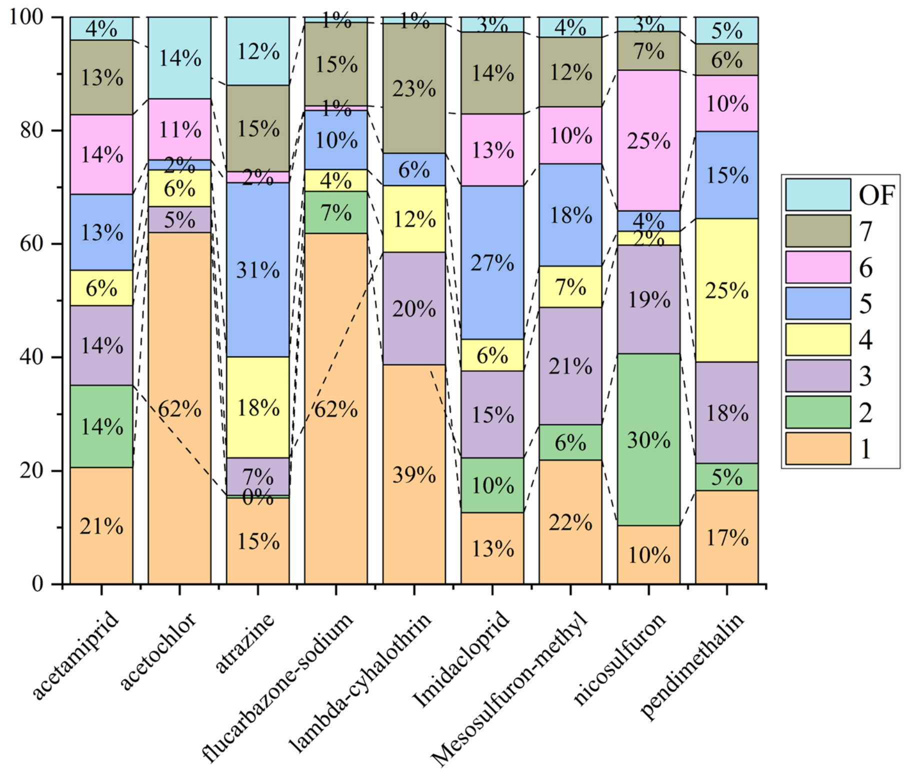

Fig. 2 Histogram of exposure concentration (EC) percentage per household for a single pesticide

The percentage of EC per pesticide in seven households and the area near the field is shown in Fig. 2.

The calculation results used 95% confidence intervals (CIs). The exposure concentration of acetamiprid was generally higher than that of other pesticides, and atrazine had the lowest EC of the nine pesticides tested. This may be related to the physical and chemical properties of pesticides, application methods, and application rates. The EC of the first household was often the highest, which could be influenced by factors such as the distance between the residence and field. Consistent with our results, imidacloprid, acetamiprid, atrazine, and pendimethalin had also been detected in previous studies at concentrations of 0.67–37 pg/m^3, 4.33–113 pg/m^3, 21.57 μg/m^3, and 31.05 μg/m^3 (Msibi et al., 2021; Zhou et al., 2020).

Figure 3 shows the lifetime non-cancer-related risks of seven households and an area near field exposed to certain pesticides in agricultural regions.

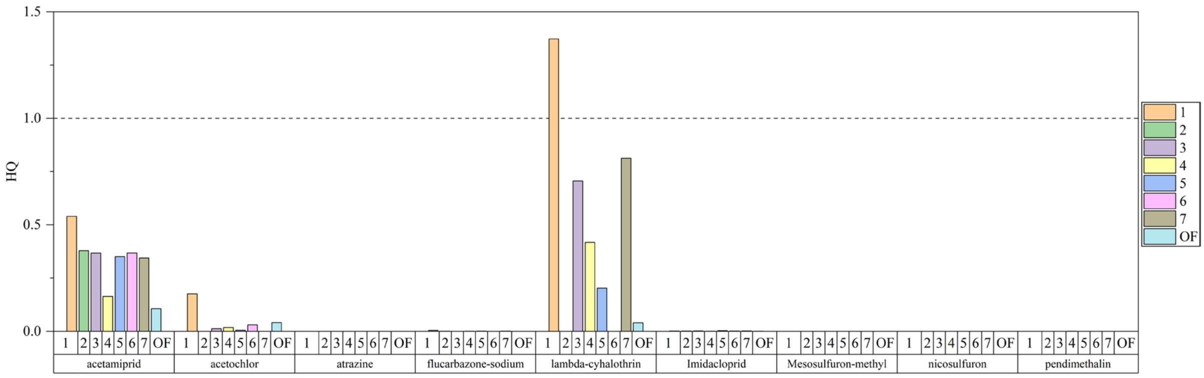

Fig. 3 Lifetime non-cancer-related risks of seven households in agricultural regions exposed to a certain pesticide

We first assessed the risk of non-occupational pesticide inhalation in seven farming households and an area around fields, the risks of chronic pesticide inhalation in grain-growing areas were acceptable for acetamiprid, acetochlor, atrazine, fluazuron-methyl, imidacloprid, methyldisulfuron-methyl, nicosulfuron-methyl, and pendimethalin. Beta cyhalothrin is neurotoxic (Shukla et al., 2017). The HQ for β-cyhalothrin in the first home was 1.3728 > 1. Since uncertainties have been factored into the reference value, which does not imply that inhabitants would become unwell from such exposure. However, a pesticide HQ > 1 nonetheless warrants a regulatory review (Hou et al., 2004).

Cancer risk assessment

We refer to the EPA's classification manual for pesticides and cancer, where two pesticides applied in this agricultural region are defined as likely to be carcinogenic to humans (EPA, 2018). Acetochlor and pendimethalin are possible human carcinogens, and their estimated lifetime cancer-related risks are shown in Fig. 4.

Exposure to pendimethalin can increase the incidence of cancer of the gastrointestinal system (Hou et al., 2004). The lifetime cancer risk of pendimethalin was lower than 1.0E−6, which means the non-occupational inhalation risk of pendimethalin for residents of seven households and nearby fields is within acceptable range. The risk of the six residential areas where acetochlor was detected was higher than 1.0E−6. Studies have shown that the use of acetochlor increases the risk of some cancers and tumors. When acetochlor is used simultaneously with atrazine, the risk of cancer is significantly increased (Lerro et al., 2015). Therefore, residents

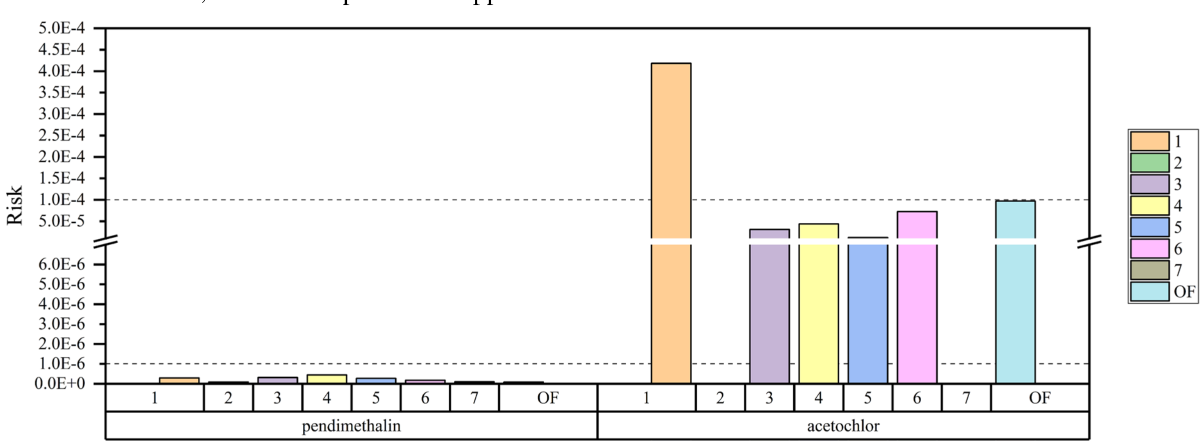

Fig. 4 Excess lifetime cancer risk of acetochlor and pendimethalin

of this agricultural area may actually face a higher risk of cancer.

Discussion

A total of 9 of the 16 pesticides investigated in the non-occupational aspiration risk assessment of pesticides were detected, all of which were currently used pesticides (CUP). Among all the detected pesticides, the pesticide with the lowest CA (contaminant concentration in air) was atrazine, and the pesticide with the highest CA was acetamiprid. The range of CA in grain-growing area is 181 pg/m^3–1.42 μg/m^3. Similar to the results of this experiment, pesticides in the concentration range of 123 ng/m^3–2.75 μg/m^3 were detected in a study in the Netherlands on the temporal and spatial changes of pesticide concentrations in indoor and outdoor air (Figueiredo et al., 2021). In a study conducted in an agricultural region of Canada, methamidophos was detected at levels ranging from 0.05 to 6.37 μg/m^3 (Garron et al., 2009).

The results showed that the indoor air concentrations of pesticides in the seven households and an area around field were generally higher than those outdoors. This may be caused by the lack of indoor air circulation and the adsorption of pesticides on indoor furniture. In addition, the personal hygiene habits of residents may also contribute to this phenomenon, such as the frequency of cleaning the room and the frequency of bathing. The risk characterization results showed that the non-cancer exposure risks of acetamiprid, acetochlor, atrazine, flazasulfuron-methyl, imidacloprid, mesosulfuron-methyl, nicosulfuron, and pendimethalin were all acceptable (HQ<1), the HQ of lambda-cyhalothrin in the first household is 1.3728 > 1, and the risk is unacceptable. The excess lifetime cancer risk in the 6 households and an area around field where acetochlor was detected was all more than 1.0E−6, requiring enhanced cancer regulatory review. Considering the effects of the remaining two factors of non-occupational exposure, residents of this agricultural area may actually be at greater risk of pesticide exposure.

The results of our study also proved that people living in agricultural areas are exposed to a variety of pesticides. And more than two types of pesticides are often present in the living environment of the inhabitants of this agricultural area at the same time. Pesticides pose more complicated health risks to humans than simply the superposition of risks. The harm to organisms increases considerably when different pesticides are combined. A mixture of pesticides commonly used worldwide can cause up to 99% mortality of larval amphibians, and the cause of this phenomenon cannot be explained by the toxicity of a particular pesticide alone (Relyea, 2009). Therefore, we only considered the effect of combined toxicity when evaluating the combination of acetochlor and atrazine due to the current lack of toxicological information. The risk of inhalation of a mixture of other pesticides cannot be determined.

The results of the assessment only represent the actual situation in both places. Since the non-occupational inhalation exposure of pesticides is affected by dozens of parameters such as application type, quantity, climate, residential location, and personal habits in each region, and there are no relevant studies for reference in China, the extrapolation data are temporarily insufficient.

Residents of agricultural areas work regularly with pesticides; therefore, there is potential for occupational exposure. Our analysis mainly assessed the dangers of indoor pesticide exposure by inhalation, not including occupational exposure. Therefore, comprehensive research is necessary to fully understand the consequences of pesticide inhalation and its potential implications on human health. The actual risk of pesticide exposure for residents in agricultural areas may be higher given the aforementioned circumstances.

To the best of our knowledge, this experiment conducted the first preliminary safety assessment of non-occupational inhalation exposure in China's grain-growing areas and established methods for the adsorption, collection, extraction, detection, and safety evaluation of pesticides in the air. However, most pesticides lack inhalation toxicological parameters, despite the vast and representative number of samples obtained (Table 1).

Conclusion

This is the first study to evaluate the possibility of pesticide exposure via inhalation among people living in a typical grain-growing region in China. We evaluated the indoor pesticide exposure of seven homes and an area around field in typical grain-growing districts of Shandong Province during spring, summer, and autumn. All nine pesticides identified in the assessment of inhalation danger were CUPs. Acetamiprid was the pesticide with the highest average EC among the nine pesticides. The first of the seven households had a higher EC. Only the first household had an estimated risk higher than the threshold in the beta-cyhalothrin non-cancer-related risk evaluation. In the acetochlor cancer risk assessment, the excess cancer risk of the first household exceeded the acceptable range, while the cancer and non-cancer-related risks of all other pesticides were within the acceptable range. We found that the levels of pesticides in indoor air were significantly higher than those outdoors.

Author contributions Yuzhao Hu (first author) helped in conceptualization, methodology, software, investigation, formal analysis, writing—original draft; Shuai Wu was involved in data curation, writing—original draft; Changcai Wu contributed to visualization, investigation; Zhimin Wei helped in resources, supervision; Jun Ning helped in software, validation; Dongmei She (corresponding author) contributed to conceptualization, funding acquisition, resources, supervision, writing—review & editing.

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Data availability Datasets are available from the corresponding author on reasonable request.

Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

Ethical approval This is an observational study. It is confirmed that no ethical approval is required.

Consent to participate Not applicable.

Consent to publish Not applicable.

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