

# Environmental Monitoring and Potential Health Risk Assessment from Pymetrozine Exposure among Communities in Typical Rice-Growing Areas of China

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## Research Article

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1 Environmental Monitoring and Potential Health Risk Assessment from  
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**Abstract:**

Pymetrozine is one of the most common insecticides used in China. This study was conducted to analyse Pymetrozine's potential exposures through various environmental routes beyond the treatment area. The aim was to estimate the potential health risk for communities due to non-dietary exposure to Pymetrozine in soil and paddy water. Data on registration of pesticides in China, government reports, questionnaires, interviews, literature reviews as well as toxicological health investigations were evaluated to determine the hazard and dose-response characteristics of Pymetrozine. These were based on the US EPA exposure and human health risk assessment methods using exposure from soil and paddy water samples collected between 10 to 20 meters around the resident's location. The potential exposures from dermal contact through soil and paddy water were estimated. The potential cancer risk from the following routes was evaluated: the ingestion through soil; dermal contact exposure through soil; dermal contact exposure through paddy water; and the potential total cancer risk for residents was less than  $1 \times 10^{-6}$ . These were within the acceptable risk levels. The potential hazard quotient (HQ) from acute and lifetime exposure by dermal contact through paddy water and soil; acute and lifetime exposure by soil ingestion for residents were less than 1, indicating an acceptable risk level, thus both potential cancer risk and hazard quotient (HQ) were relatively low. Potential human health risk assessment of Pymetrozine in soil and paddy water suggested that negligible cancer risk and non-cancer risk based on ingestion and dermal contact are the main potential routes of exposure to residents.

**Keywords:** agriculture communities, China, environmental routes, health risk, Pymetrozine

58

**59 1. Introduction**

60 Pesticides play a significant role in reducing the losses of agricultural products and improve  
61 the affordable yield and quality of food (Aktar et al., 2009, Schreinemachers et al., 2017).

62 Pesticides can be toxic to other organisms, including birds, fish, beneficial insects, and non-  
63 target plants, as well as contaminate air, water, soil, and crops (Tudi et al., 2021). Moreover,  
64 pesticide contamination can move away from the application areas resulting in environmental  
65 pollution. Such chemical residues impact human health through environmental and food  
66 contamination (Yadav et al., 2015).

67

68 Although some measures have been proposed to reduce the negative effects of pesticides on  
69 the environment and human health (Phung et al., 2012c), both acute and chronic human  
70 toxicities resulting from these substances remain a serious problem (Tudi et al., 2021). It is  
71 predicted that the high-risk of pesticide exposure will increase worldwide over the next decade,  
72 especially in developing countries (Delcour et al., 2015). China leads the world in both  
73 pesticide production and consumption (Zhang et al., 2011). There was an increase in the use of  
74 pesticides from 1.28 million tons in 2000 to 1.8 million tons in 2013, with an average annual  
75 increase of 2.7% reported for 2018 (Shuqin and Fang, 2018).

76

77 It has been reported that 70% of the pesticides used in China were not absorbed by plants and  
78 other organisms, instead they have entered into the soil and groundwater (Wang et al.,  
79 2018). Thus, many non-farmworker residents living close to agricultural lands where  
80 pesticides are often used intensively are exposed to pesticides. Owing to the intrinsic toxicity  
81 of pesticides, it is important to evaluate the potential health consequences for this specific  
82 population, but such evaluations are largely absent in studies to date.

83

84 Pymetrozine {4, 5-dihydro-6-methyl-4-[(3-pyridylmethylene)-amino]-1, 2, 4-triazine-3(2H)-  
85 one} which has the basic structure of a pyridine azomethine (Zhang et al., 2015). Currently, is  
86 widely used in China (Jia et al., 2019, Gong et al., 2019, Kovacova et al., 2013). In addition,  
87 Pymetrozine has recently replaced organic phosphate pesticides although organophosphate  
88 pesticides continue to be used in some other countries in the world (Hamsan et al., 2017, Atabila  
89 et al., 2018, Phung et al., 2012a ). But little is known of the potential risk posed to residents  
90 on adjoining land, from using Pymetrozine. A first step in the development of a potential health  
91 risk assessment in respect to this chemical must be to gain an understanding of its  
92 environmental distribution, attenuation, and fate under field conditions.

93

94 Pymetrozine has a negative influence on reproduction, causes irritation to the respiratory tract  
95 (European Food Safety, 2017) Furthermore, Pymetrozine has brought great benefits to crop  
96 production by killing insects. The United States Environmental Protection Agency (USEPA)  
97 classifies Pymetrozine as a possible human carcinogen and a cancer slope factor of is 0.0019  
98 mg/kg quoted by USEPA (2010), and this will be further considered in the present evaluation.  
99 However, most previous studies of these parameters were not carried out under field conditions  
100 (Europe Food safety Authority, 2014).

101

102 This study was conducted to analyse Pymetrozine potential exposures through the  
103 environmental routes for residents living close to agricultural lands in typical rice growing  
104 areas of China, to estimate potential health risk for residents due to non-dietary exposure to  
105 Pymetrozine in soil and paddy water. This study will provide scientific information for policy  
106 makers to set the proper pesticide application techniques and methods to minimize the pesticide  
107 exposure relating to health effects for communities.

108

## 109 **2. Study Area**

110 The study area was identified with the assistance of the Institute of Plant Protection Chinese  
111 Academy of Agricultural Science and Beijing ECO-SAF Technology Co., Ltd. Based on the  
112 location of the Institute of Plant Protection Chinese Academy of Agricultural Science and  
113 Beijing ECO-SAF Technology Co., Ltd and local government, the typical rice-growing areas  
114 of Hunan and Guangxi provinces were selected as the study areas.

115

116 In the present study, the Yun-Wen Village Shang Lin Country Nan Ning City Guangxi  
117 Province and Hua Tang Village Chang Sha Country Chang Sha City Hunan Province were the  
118 study areas (Figure 1). Samples of paddy soil and paddy water were collected in association  
119 with a spraying event. Farmers sprayed pesticides once a month. Residents lived within 10-20  
120 meters of their agriculture areas.

121

## 122 **3. Materials and Methods**

### 123 **3.1 Sample Collection**

124 The quartering sampling method (Li, 2008) was used to collect soil and paddy water samples.  
125 Four samples were collected from the surface (0-15 cm layer) in every plot (size about 2000  
126 m<sup>2</sup>), composited and stored in a polyethylene bag as one sample. Fifteen soil and fifteen paddy  
127 water samples were collected in each region. Samples were collected on the day prior to  
128 spraying, the day of spraying insecticides and 1, 3, 5, 7, 9, 14, 21 and 28-days after insecticide  
129 spraying. All the samples were stored at -20°C immediately prior to analysis and the samples  
130 were analysed within the one month.

131

### 132 **3.2 Sample Analysis**

133 Based on the previous study (Li et al., 2011), the modified QuEChERS method was adapted to  
134 analyse both of the soil and paddy water samples. The parameters including linearity, linear  
135 range, LOQs, accuracy, precision and stability were considered to evaluate the method  
136 validation. The result indicated that the analytical method of Pymetrozine was accuracy and  
137 precision and it meets the requirement.

138

### 139 **3.3 Exposure Assessment through Environmental Media**

140 Pymetrozine has only recently replaced organophosphate insecticides, thus there are very few  
141 studies regarding the exposure of operators and the community. Ingestion of vegetables and  
142 fruits is considered as the main exposure route of Pymetrozine in previous studies conducted  
143 in China (Jia et al., 2019, Gong et al., 2019, Kovacova et al., 2013, Yu et al., 2020). However,  
144 there is little study focus on the Pymetrozine exposure from soil and paddy water and its  
145 environmental health risk assessment for the agriculture communities in China.

146

147 In this study, the soil and paddy water samples collected within 10 -20 meters of the residents'  
148 apartments and two main potential pathways of the exposure assessment were considered.  
149 Dermal contact and ingestion are the main exposure routes of communities exposed to  
150 Pymetrozine in soil and paddy water in the study areas.

151

152 Developing a conceptual site model (CSM) can assist the process of understanding how human  
153 'receptors' may be exposed to chemicals from relevant environmental sources. The CSM  
154 describes the sources of contamination, the pathways by which contaminants may migrate  
155 through the various environmental media and the populations that may potentially be exposed.  
156 CSMs are particularly important in environmental health risk assessment of contaminated sites  
157 and based on these aspects, the conceptual framework was settled in this study (Figure 2).



158

### 159 3.3.1 Calculation of Absorbed Daily Dose (ADD) of Pymetrozine from Dermal Exposure

160 The Absorbed Daily Dose (ADD) of Pymetrozine from dermal exposure in one spraying event  
161 was estimated using the following equation:

$$162 \quad \text{ADD dermal} = \frac{C * SA * SL * ABS}{BW} * 10^{-6} \quad (1)$$

163

164 Where, ADD is the average daily dose via dermal contact;. *C* is the concentration of  
165 pymetrozine in soil or water sample ( $\mu\text{g}/\text{kg}$ ); *BW* is body weight (kg); *SA* is the exposed skin  
166 area ( $\text{m}^2$ ), reported by the Environmental Ministry of China (2010); *SL* is the skin adherence  
167 factor; and *ABS* is the dermal absorption factor for pymetrozine (US EPA, 2000) (TableS1).

168

### 169 3.3.2 Calculation of Absorbed Daily Dose (ADD ingestion) Pymetrozine from Ingestion 170 Exposure

171 ADD of pymetrozine from ingestion exposure in one spraying event was estimated using the  
172 following equation:

$$173 \quad \text{ADD ingestion} = \frac{C * IngR}{BW} * 10^{-6} \quad (2)$$

174

175

176 Where, ADD ingestion is the average daily doses of via ingestion; *C* is the concentration of  
177 pymetrozine in soil sample( $\mu\text{g}/\text{kg}$ ); *IngR* is the ingestion rate for pymetrozine (US EPA, 2000);  
178 and *BW* is body weight (Environmental Ministry of China, 2010) (Table S1).

179

### 180 3.3.3 Total Acute Dermal and Ingestion Exposure Dose of Pymetrozine with Communities

181 In this section, based on the results of the acute ingestion exposure level of Pymetrozine from  
 182 soil (on the day insecticide spraying, 1, 3, 5, 7, 9, 14, and 21-day after insecticide spraying in one  
 183 spraying event separately), the acute dermal exposure level of Pymetrozine from soil (on the  
 184 day of insecticide spraying, 1, 3, 5, 7, 9, 14, and 21-day after insecticide spraying in one spraying  
 185 event separately) and the acute dermal exposure level of Pymetrozine from paddy water (on  
 186 the day insecticide spraying, 1, 3, 5, 7, 9 and 14-day after insecticide spraying in one spraying  
 187 event separately) were calculated, and then sum of the post-application exposure dose to  
 188 calculate the total acute dermal exposure level of Pymetrozine from soil, the total acute dermal  
 189 exposure level of Pymetrozine from paddy water; and the total acute ingestion exposure level  
 190 of Pymetrozine from soil separately.

191

### 192 **3.3.4 Calculation of Lifetime Average Daily Dose (LADD) of Pymetrozine from Dermal** 193 **and Ingestion Exposure**

194 The Lifetime Average Daily Doses (LADD) of Pymetrozine from dermal exposure and  
 195 ingestion by the communities were estimated using the following equations:

$$196 \quad \text{LADD dermal} = (\text{ADD (dermal)} \times \text{EF} \times \text{ED}) / \text{AT} \quad (3)$$

197

198

$$199 \quad \text{LADD ingestion} = (\text{ADD (ingestion)} \times \text{EF} \times \text{ED}) / \text{AT} \quad (4)$$

200

201 Where, ADD (mg/kg/day) is the sum of dermal absorbed daily dose and the ingestion absorbed  
 202 daily dose of Pymetrozine of the communities; LADD is the lifetime average daily dose of  
 203 Pymetrozine; EF is the exposure frequency (number of days per year); there are four spraying  
 204 times in this study area and the concentration level of the Pymetrozine in soil was detected

205 from the day insecticide spraying until after 21-day insecticide spraying; and the concentration  
 206 level of Pymetrozine in paddy water was detected from the day insecticide spraying until after  
 207 14-day insecticide spraying, thus the frequencies of exposure for soil and paddy water were 84  
 208 days and 56days separately. ED is the exposure duration (lifetime years) and the exposure  
 209 duration for adults is 70 years. AT is the life expectancy in years and the life expectancy for  
 210 adults is (365\*70). (Table S1).

211

### 212 **3.4 Potential Environmental Health Risk Characterization**

213 The potential health risks caused by main routes of exposure to chemical contaminants in  
 214 environment were assessed based on the health risks models of the U.S. EPA (2004).

215

#### 216 **3.4.1 Non-carcinogenic hazard quotient (HQ)**

217 The non-cancer risks of exposure to Pymetrozine in soil and paddy water were calculated as  
 218 follows (Eqs. (5)- (6):

$$219 \quad HQ = \frac{LADD}{RFD} \quad (5)$$

220

$$221 \quad HI = \sum HQ \quad (6)$$

222

223 As stated earlier, Acute and Lifetime Average Daily Dose (LADD) of Pymetrozine in soils and  
 224 paddy water through multiple pathways were calculated using Eqs. (1)- (4) Separately. The  
 225 hazard quotient (HQ) represents the non-carcinogenic risk for Pymetrozine through different  
 226 exposure pathways. The reference doses (*RFD*) were taken from the U. S. Department of  
 227 Energy's RAIS compilation (U. S. Department of Energy 2000 and 2010). The hazard index

228 (HI) represents the total non-carcinogenic risk for Pymetrozine through different exposure  
229 pathways.

230

### 231 **3.4.2 Carcinogenic risk**

232 The carcinogenic risk (CR) is the incremental probability of an individual developing cancer  
233 over a lifetime due to carcinogenic exposure. The carcinogenic risk is evaluated by Eq. (7):

$$234 \quad CR = LADD \times CSF \quad (7)$$

235 The estimated *CR* is the probability of an individual developing in any type of cancer from  
236 lifetime exposure to carcinogenic hazards. (*LADD*) is the lifetime average daily dose of  
237 Pymetrozine and *CSF* is the cancer slope factor, and it is expressed in  $(\mu\text{g}/\text{kg}/\text{day})^{-1}$ .

238

239 To evaluate the total potential cancer risks (TCR) from different pathways for soil and paddy  
240 water, these parameters were calculated using Eq. (8).

$$241 \quad TCR = \sum^n CR \quad (8)$$

242 *CR* is the individual carcinogenic risk of every pathway; *n* is different pathways that cause  
243 cancer risk.

244

## 245 **4. Results and Discussion**

### 246 **4.1 Hazard Identification of Pymetrozine**

247 The identification of Pymetrozine as the major hazard to community's health was carried out  
248 by using data on registration of pesticide in China, government report, questionnaire and  
249 interview, and literature reviews as well as toxicological health investigation.

250

#### 251 **4.1.1 Pymetrozine Formulations Used in China**

252 In China, since 2008, because Pymetrozine is being employed as a main substitute for  
253 pesticides of high toxicity, there exists a need to properly quantitate the human health risks that  
254 result from the used of Pymetrozine in rice paddies (Wu, et al, 2013; Jiang Su government  
255 report, 2017).

256

#### 257 **4.1.2 Human Health Effects of Pymetrozine Usage in China**

258 Residents are exposed to the residue of pesticides through environmental media such as soil,  
259 water and food by different routes of exposure including inhalation, ingestion and dermal  
260 contact; and that lead to acute and chronic diseases (Damalas and Eleftherohorinos, 2011).

261

262 From the survey and interview in our study (Table S2), it could be found that most of the  
263 farmers use the locally made spraying equipment which doesn't have appropriate safeguards.  
264 As a result of substandard construction of the equipment, cracks and leaks can happen easily  
265 during the application. In addition, applicators have either inadequate or no protective clothing,  
266 masks, and gloves. Furthermore, owing to the lack of proper instruction and training farmers  
267 do not identify the harmful pests from the other non-harmful pests and use the incorrect nozzles.  
268 The spraying equipment which farmers use is not properly cleaned and handled when they  
269 finish the spraying. Therefore, owing to the spills and splashes, direct spray contact, or even  
270 drift during the pesticide application under the improper way, agriculture workers and residents  
271 living around the agriculture land are potentially exposed to pesticides via dermal contact,  
272 ingestion and respiratory inhalation (Sugeng et al., 2013).

273

274 It was observed that there are many discarded pesticide packaging bags in the study areas. After  
275 pesticides are used to target plants, they will go through transfer/migration and degradation in  
276 the environment (Tudi et al., 2021). Improper use of pesticide, its management and behavior

277 also lead to environmental pollution (Connell, 2018) including soil, water, air, and food  
278 contamination. The residues of pesticides from these different environmental medias may enter  
279 the human body and result in negative impact on human health (Tudi et al., 2021).

280

281 Currently, there are very few studies related to the health effects of Pymetrozine in China and  
282 other countries. Previous studies indicate that acute toxic effects always occur from within a  
283 few minutes to several hours after poisoning by pesticides (Yang and Deng, 2007, DeBleecker,  
284 1995, Pereira et al., 2015, Atabila et al., 2018b). Also, the documented issues indicate that  
285 various chronic diseases and disorders sometimes occur after people have been exposed to  
286 pesticides (Wesseling et al., 1997, Uram, 1989, Phung et al., 2012b). Therefore, there may be  
287 exposure and potential health effects from Pymetrozine application by the occupational,  
288 environmental and dietary routes to both agriculture workers and residents who are living  
289 around the agriculture areas.

290

## 291 **4.2 Potential Exposure Assessment of Pymetrozine through Environmental Media**

### 292 **4.2.1 Baseline Exposure Levels of Pymetrozine through Environment with Agriculture**

#### 293 **Communities**

294 Soil and paddy water were collected one-day prior to application, and the concentration level  
295 of Pymetrozine in soil and paddy water were analysed. The Pymetrozine concentration level in  
296 soil and paddy water which were collected one-day prior to application is under the detection,  
297 thus the baseline exposure level of Pymetrozine in the environment was set to be zero.

298

### 299 **4.2.2 Potential Acute Dermal and Ingestion Exposure Levels of Pymetrozine in** 300 **Communities**

301 Based on the concentration level of Pymetrozine in soil and paddy water and the US EPA (2004)  
302 exposure assessment method, the potential acute dermal exposure level of Pymetrozine in soil  
303 and paddy water and the potential acute ingestion exposure level of Pymetrozine in soil were  
304 calculated for each day of each sampling site during one spraying event (day of insecticide  
305 spraying, 1-day after insecticide spraying, 3-day after insecticide spraying, 5-day after  
306 insecticide spraying, 7-day after insecticide spraying, 9-day after insecticide spraying, 14-day  
307 after insecticide spraying and 21-day after insecticide spraying).

308

309 The concentration level of Pymetrozine on the day of insecticide spraying varies in different  
310 agriculture lands. The application rate by different farmers, the target areas of the sprayed  
311 insecticide and the kind of equipment used by different farmers are different, and these are the  
312 main reasons caused the differences in the initial concentration level of Pymetrozine in the soil  
313 and the exposure level of Pymetrozine through the soil (Li, 2010). In addition, the previous  
314 study also shows that the initial concentration level of Pymetrozine is related to the difference  
315 in sampling strategy, the difference in climate and soil characteristic, the density of rice  
316 planting and the difference in growth trends (Yang, 2011). These are further main reasons for  
317 the difference in Pymetrozine acute dermal exposure level and acute ingestion exposure level  
318 through soil in different agriculture lands in both Hunan and Guangxi areas.

319

320 The potential minimum, mean, 95<sup>th</sup> percentile, and maximum values of the potential acute  
321 dermal exposure through soil and paddy water, and potential acute ingestion exposure through  
322 soil for adults in Guangxi and Hunan after Pymetrozine application are plotted in TableS3, S4  
323 and S5, separately. The result indicates that adults were exposed to Pymetrozine from the day  
324 of insecticide spraying until 21 days after insecticide spraying. The potential acute dermal  
325 exposure through soil was higher than acute dermal exposure through paddy water in both

326 Guangxi and Hunan areas. The potential acute dermal exposure level through soil; the potential  
327 acute dermal exposure level through paddy water were lower than the potential acute ingestion  
328 exposure level through soil for adults in both areas separately.

329

330 Table S3 indicate that the potential acute dermal exposure level for adults through soil in Hunan  
331 on the day of Pymetrozine application was increased to  $1.2 \times 10^{-6} \mu\text{g}/\text{kg}/\text{day}$  which was  $1.2$   
332  $\times 10^{-6}$ -fold higher than the baseline exposure level. The potential exposure level decreased to  
333  $0.02 \times 10^{-6} \mu\text{g}/\text{kg}/\text{day}$  after 21 days of insecticide spraying. The potential acute dermal  
334 exposure level for adults through soil in Guangxi on the day of Pymetrozine application  
335 increased to  $1.1 \times 10^{-6} \mu\text{g}/\text{kg}/\text{day}$  which was  $1.1 \times 10^{-6}$ -fold higher than the baseline exposure  
336 level. The potential exposure level decreased to  $0.02 \times 10^{-6} \mu\text{g}/\text{kg}/\text{day}$  after 21-days of  
337 insecticide spraying.

338

339 Table S4 indicate that the potential acute dermal exposure level for adults through paddy water  
340 in Hunan on the day of Pymetrozine application increased to  $0.5 \times 10^{-6} \mu\text{g}/\text{kg}/\text{day}$  which was  
341  $0.5 \times 10^{-6}$ -fold higher than the baseline exposure level. The potential exposure level decreased  
342 to  $0.04 \times 10^{-6} \mu\text{g}/\text{kg}/\text{day}$  after 14 days of insecticide spraying. The potential acute dermal  
343 exposure level for adults through paddy water in Guangxi on the day of Pymetrozine  
344 application increased to  $0.6 \times 10^{-6} \mu\text{g}/\text{kg}/\text{day}$  which was  $0.6 \times 10^{-6}$ -fold higher than the  
345 baseline exposure level. The potential exposure level decreased to  $0.04 \times 10^{-6} \mu\text{g}/\text{kg}/\text{day}$  after  
346 14 days of insecticide spraying.

347

348

349 Table S5 indicate that the acute ingestion exposure level for adults through soil in Hunan on  
350 the day of Pymetrozine application increased to  $1.02 \times 10^{-4} \mu\text{g}/\text{kg}/\text{day}$  which was  $1.02 \times 10^{-4}$   
351 -fold higher than the baseline exposure level. The potential exposure level decreased to  $1.73$   
352  $\times 10^{-6} \mu\text{g}/\text{kg}/\text{day}$  after 21 days of insecticide spraying. The potential exposure level decreased



353 to  $4.18 \times 10^{-6} \mu\text{g}/\text{kg}/\text{day}$  after 21 days of insecticide spraying. The potential acute ingestion  
354 exposure level for adults through soil in Guangxi on the day of Pymetrozine application  
355 increased to  $7.64 \times 10^{-5} \mu\text{g}/\text{kg}/\text{day}$  which was  $7.64 \times 10^{-5}$ -fold higher than the baseline  
356 exposure level. The potential exposure level decreased to  $5.439 \times 10^{-5} \mu\text{g}/\text{kg}/\text{day}$  after 14 days  
357 of insecticide spraying.

358

#### 359 **4.2.3 Potential Total Acute Exposure of Pymetrozine in Communities**

360 The descriptive analysis was carried out on the total acute dermal exposure level of  
361 Pymetrozine through soil, the total acute ingestion exposure level of Pymetrozine through soil,  
362 the total acute dermal exposure level of Pymetrozine through paddy water in Guangxi and  
363 Hunan. The results are shown in Table S6. It shows that there is no significant difference  
364 between the total dermal exposure from soil ( on the day insecticide spraying, 1, 3,5,7, 9,14  
365 and 21-day after insecticide spraying) and the acute soil dermal exposure on the day insecticide  
366 spraying; there is no significant difference between the total dermal exposure from paddy water  
367 and the acute dermal exposure from paddy water on the day insecticide spraying; and there is  
368 no significant difference between the total ingestion exposure level from soil and the acute  
369 ingestion exposure level from soil on the day insecticide spraying for adults in Guangxi and  
370 Hunan.

371

#### 372 **4.2.4 Potential Lifetime Average Daily Dose (LADD)**

373 Based on the potential total acute dermal exposure levels of Pymetrozine through soil; potential  
374 total acute ingestion exposure levels of Pymetrozine through soil; and the potential total acute  
375 dermal exposure levels of Pymetrozine through paddy water during the one spraying event, the  
376 potential life-time exposure levels of Pymetrozine in soil and paddy water were calculated for  
377 each sampling point. The results of the descriptive analysis are indicated in Table S7. The

378 LADD for Pymetrozine exposure with agriculture communities assumed that the farmers  
379 sprayed the same amount of Pymetrozine for every spray event and communities are exposure  
380 to Pymetrozine in short-time period in every year.

381

### 382 **4.3 Potential Environmental Risk Characterization of Pymetrozine exposure for** 383 **agriculture communities**

384 The potential cancer risk assessment of Pymetrozine through dermal contact, and ingestion  
385 exposure routes for adults, as well as the exposure doses of Pymetrozine in relation to adults'  
386 potential non-carcinogenic risk through dermal contact and ingestion exposure routes in Hunan  
387 and Guangxi were calculated using the parameters of the US EPA health risk assessment  
388 method in this study.

389

390 Since there are no human studies about the Pymetrozine, to make summaries derived from the  
391 results of sub-chronic and chronic toxicity, metabolism, and dermal penetration studies in  
392 animals, these studies indicate that Pymetrozine impacts on three major organs in the body  
393 including liver, hematopoietic system, and lymphatic system. In addition, from both sub-  
394 chronic and chronic studies from dogs, it is found that this chemical affects muscle tissue.  
395 Pymetrozine has been reported that it has significant adverse health effects on mice, rats, and  
396 dogs (US EPA, 2000). For example, hepatocellular hypertrophy is related to induction of drug  
397 metabolizing enzymes (USEPA, 2010). The EPA also classifies Pymetrozine as a possible  
398 human carcinogen (USEPA, 2010).

399

400 According to the available slope factors discussed by the US EPA (2010), dermal contact and  
401 ingestion were involved in the risk estimation of Pymetrozine. The cancer slop factor is 0.0019  
402 mg/kg. The value of the Chronic Population-Adjusted Dose from Pymetrozine for general

403 population is 0.0038 mg/kg/day that was used to calculate the non-cancer risk for lifetime  
404 exposure dose. The value of the Acute Population-Adjusted Dose from Pymetrozine for general  
405 population is 0.42 mg/kg and that was used to calculate the non-cancer risk for acute exposure  
406 dose.

407

#### 408 **4.3.1 Potential Cancer Risk**

409 The distribution of potential cancer risk is presented in Figure 3 and Table S8. The potential  
410 cancer risk of dermal contact through soil for adults in both areas were higher than the cancer  
411 risk of dermal contact through paddy water. The potential cancer risk of dermal contact through  
412 soil for adults in both areas were lower than the potential cancer risk of ingestion through soil  
413 in both areas.

414

415 In generally, the minimum, average, 95<sup>th</sup> percentile, and maximum of the potential cancer risk  
416 of dermal contact through soil and water for adults; the minimum, average, 95<sup>th</sup> percentile and  
417 maximum of the potential cancer risk of ingestion through soil for adults; the potential total  
418 cancer risk through soil and paddy water in both areas were less than  $1 \times 10^{-6}$ , being within the  
419 acceptable level.

420

#### 421 **4.3.2 Potential Non-cancer Risk from Lifetime Exposure Dose and Acute Exposure Dose**

422 The distribution of the potential non-cancer risk from both lifetime and acute exposure dose  
423 are presented in Fig. 4 and Table S9, and Fig 5 and Table S10.

424

425 The results indicate that the minimum, average, 95<sup>th</sup> percentile and maximum of the potential  
426 non-cancer risk from both of lifetime and acute dermal contact exposure dose through soils;  
427 the minimum, average, 95<sup>th</sup> percentile and maximum of the potential non-cancer risk from

428 both lifetime and acute dermal contact exposure dose through paddy water; the minimum,  
429 average, 95<sup>th</sup> percentile and maximum of the potential non-cancer risk from the both lifetime  
430 and acute ingestion exposure dose through soil for adult in two study areas were below 1. The  
431 minimum, average, 95<sup>th</sup> percentile and maximum of the potential non-cancer risk from both  
432 lifetime and acute dermal contact exposure dose through soils; the minimum, average, 95<sup>th</sup>  
433 percentile and maximum of the potential non-cancer risk from the lifetime and acute dermal  
434 contact exposure dose through paddy water; the minimum, average, 95<sup>th</sup> percentile and  
435 maximum of the potential non-cancer risk from both lifetime and acute ingestion exposure dose  
436 through soil for adults of these two areas below 1. The potential total non-cancer risk from both  
437 lifetime exposure dose and acute exposure dose through paddy water and soil in these two areas  
438 were less than 1. Based on the US EPA (2010) report, if  $HI < 1$ , the exposed individual was  
439 unlikely to obviously experience adverse health effects. On the contrary, if  $HI > 1$ , there was a  
440 chance of non-carcinogenic effect. Thus, the potential non-cancer risk from Pymetrozine  
441 through both soil and paddy water were relatively low, indicating an acceptable risk level.

442

443 The results also indicated that the potential non-cancer risk from both lifetime and acute dermal  
444 contact dose through soil for adults in both areas were higher than potential non-cancer risk  
445 from both lifetime and acute dermal contact dose through paddy water separately. The potential  
446 non-cancer risk from both lifetime and acute dermal contact exposure dose through soil for  
447 adults in both areas were lower than the potential non-cancer risk from both lifetime and acute  
448 ingestion exposure dose through soil in both areas and these results are consistent with the  
449 previous results (Bhandari et al., 2020, Landrigan and Goldman, 2011, Pan et al., 2018, Yadav  
450 et al., 2016).

#### 451 **4.4 Sensitivity Analysis**

452 The percentage contribution of exposure pathways is used to determine which variables and  
 453 pathways most strongly influence the potential risk estimate (Tudi et al., 2019). Percentage  
 454 contribution of exposure pathways to potential total risk is calculated by the following equation:

455

$$HI_{total} = \sum_{i=1}^n (HI_i)$$

456

457

(Eq 6)

458

$$\text{Percent contribution}_i = \frac{HI_i}{HI_{total}} * 100\%$$

459

(Eq 7)

460 The result indicated that in both areas, the risk index from ingestion exposure dose from  
 461 Pymetrozine through soil took up the highest percentage in the potential total cancer risk  
 462 (98.46%) and the potential total non-cancer risk (98.28%), followed by the potential risk index  
 463 of dermal contact exposure dose from Pymetrozine through soil and then the potential risk  
 464 index of dermal contact exposure dose from Pymetrozine through paddy water (Table 1)

#### 465 **4.5 Uncertainty Analysis**

466 The following uncertainties limit the validity of the results of this research work. (1) the  
 467 concentrations of pesticides across the different rice growing seasons are different (Zhu et al.,  
 468 2017), but seasonal variation of pesticides were not investigated. Thus, the exposure and  
 469 potential risk level of Pymetrozine may not present the real situation in this study. (2) CSF and  
 470 RFD were treated as constants for all members of population, but they should be different from  
 471 person to person (Phung et al., 2012a, Phung et al., 2013). In this study, each input parameter  
 472 is considered by a point estimate. Variability and uncertainty should be considered when  
 473 choosing the input (Sadler et al., 2016). The traditional way of managing uncertainty and  
 474 variability has been to incorporate safety factors or use conservative assumptions, which can

475 lead to unrealistically high estimations which are neither transparent nor efficient when further  
476 testing or measures might be necessary (Atabila, 2017). Conversely, it is also possible to  
477 underestimate the exposure for sensitive populations. Deterministic estimations cannot explain  
478 the number of individuals that might be exposed to a dose over a reference value or the  
479 probability of a certain exposure (Atabila et al., 2018a). Furthermore, deterministic estimations  
480 are given with a precision that does not reflect the uncertainty and variability that is inevitable  
481 in such assessments. However, variability and uncertainty are not considered or evaluated in  
482 the calculations of this study. (3) The exposure factors for the body weight and life time  
483 frequency as constants for all members of population, but they could vary from person to person;  
484 the data analyses showed that there is a major inter individual variability inherent in these  
485 exposure factors, which must be considered in risk assessments (Tudi et al., 2019). However,  
486 there is not only variability between individuals within the same gender and age group, but also  
487 between different age groups and between females and males (Filipsson, 2011). However, this  
488 study does not consider this aspect. (4) the concentrations of parent material of Pymetrozine  
489 found in the water and soil samples were considered as a main hazardous source and residents  
490 were mainly exposed to it but the metabolism products of the Pymetrozine in both soil and  
491 paddy water could also pose risk to human (US EPA, 2010). However, the potential risk of the  
492 metabolism products of Pymetrozine from soil and paddy water was not considered in this  
493 study, thus it somewhat underestimated the level of both cancer and non-cancer risk assessment.  
494

## 495 **5. Conclusions**

496 The potential acute dermal exposure levels of Pymetrozine through soil and paddy water, acute  
497 ingestion exposure level of Pymetrozine through soil, total acute dermal exposure levels of  
498 Pymetrozine through soil, total acute dermal exposure levels of Pymetrozine through paddy  
499 water, and potential total acute ingestion exposure levels of Pymetrozine through soil peaked

500 at the day of insecticide application and returned to the normal range approximately 21-days  
501 after insecticide application.

502

503 The potential total dermal exposure and ingestions exposure levels of Pymetrozine through soil  
504 and paddy water were slightly different from the potential acute dermal exposure levels on the  
505 day of insecticide spraying due to the small contribution after 1, 3, 5, 7, 9, 14 and 21-day  
506 exposure levels to the ADDT.

507

508 For both areas, the main exposure routes for potential carcinogenic and potential non-  
509 carcinogenic risks of Pymetrozine were the soil ingestion, followed by the soil dermal contact  
510 and then the paddy water dermal contact. Thus, the soil ingestion route caused the highest  
511 exposure of the potential carcinogenic risk and non-carcinogenic risk to human health.

512

513 In general, the results of potential risk characterization indicate that agriculture communities  
514 have a relatively low potential risk of adverse health effects from the Pymetrozine application  
515 in rice growing areas.

516 Authors' contributions MT: conceptualization, data curation, formal analysis, methodology,  
517 validation, writing—original draft, and writing—review and editing. RDH: validation,  
518 resources, supervision, funding acquisition, and writing—review and editing. ST: validation,  
519 resources, supervision. LW: validation, resources, supervision. AA: validation, resources,  
520 supervision: RS: validation, resources, supervision; DC: validation, resources, supervision;  
521 DTP: validation, resources, and project administration'

522

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524

525 **Data availability:** The datasets used and/or analysed during the current study are available  
526 from the corresponding author on reasonable request.

527 **Compliance with ethical standards**

528 **Conflict of interest**

529 The authors declare that they have no conflict of interest, financial or other. This article does  
530 not contain any studies involving human participants or animals.

531 **Ethical approval.** Not applicable

532 **Consent to participate.** Not applicable

533 **Consent to publish** Not applicable

534

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- 660

# Figures

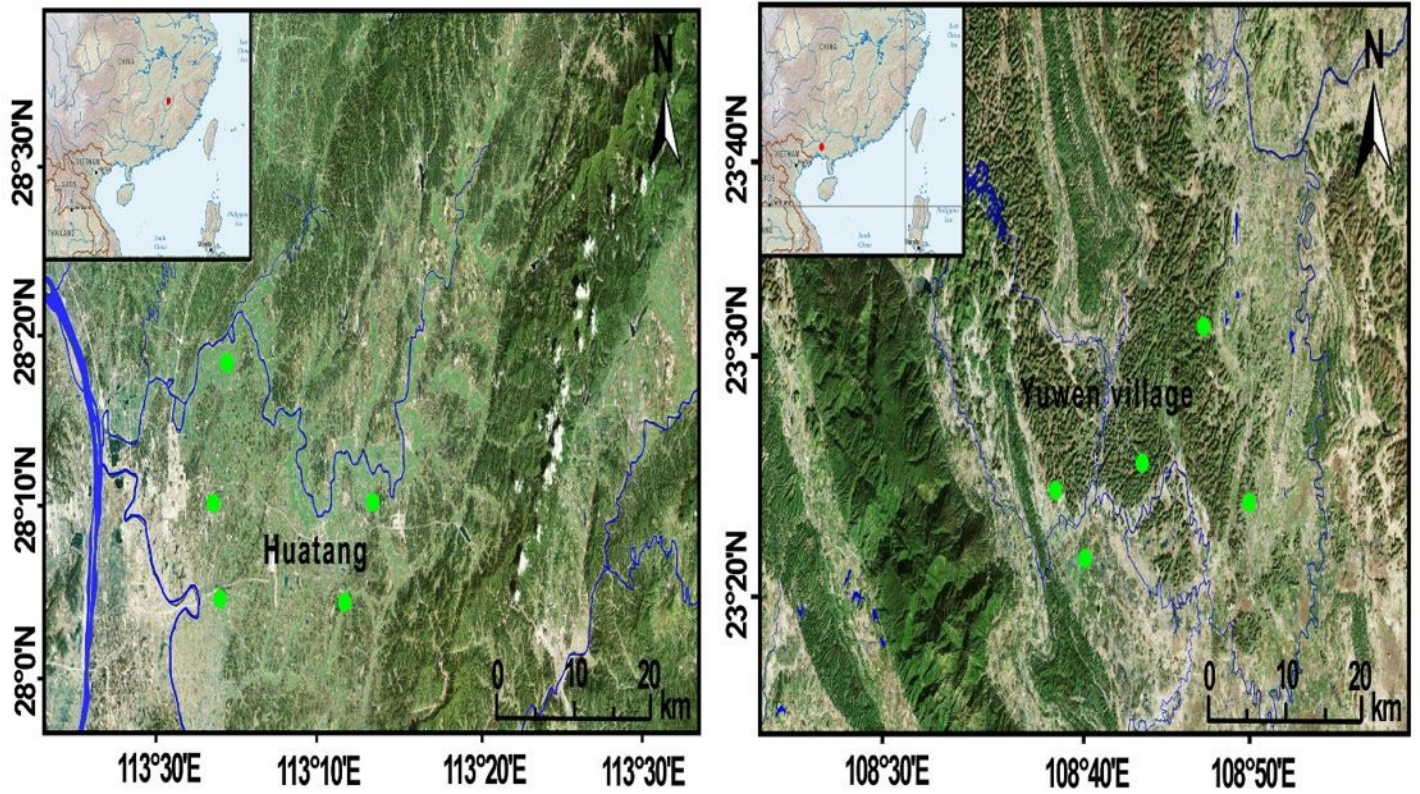


Figure 1

Location Map of the Sampling Sites

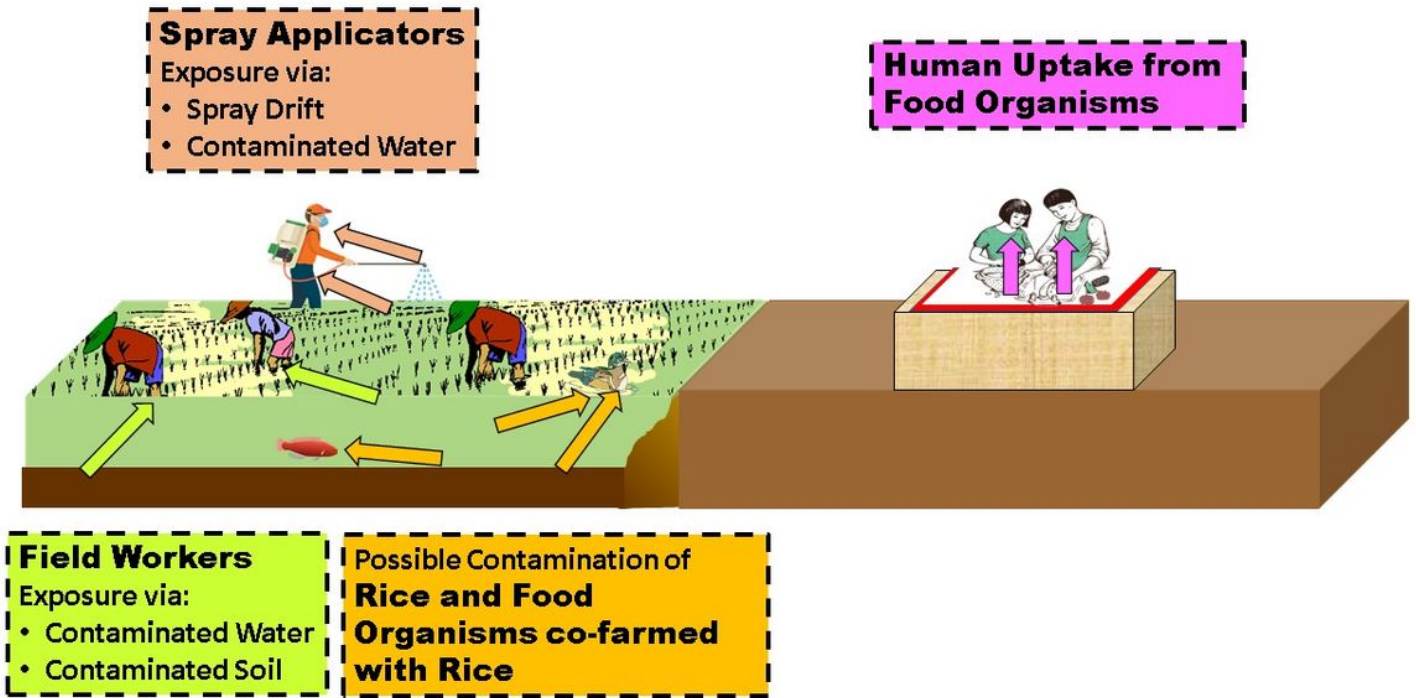


Figure 2

Conceptual Diagram of the Exposure Routes

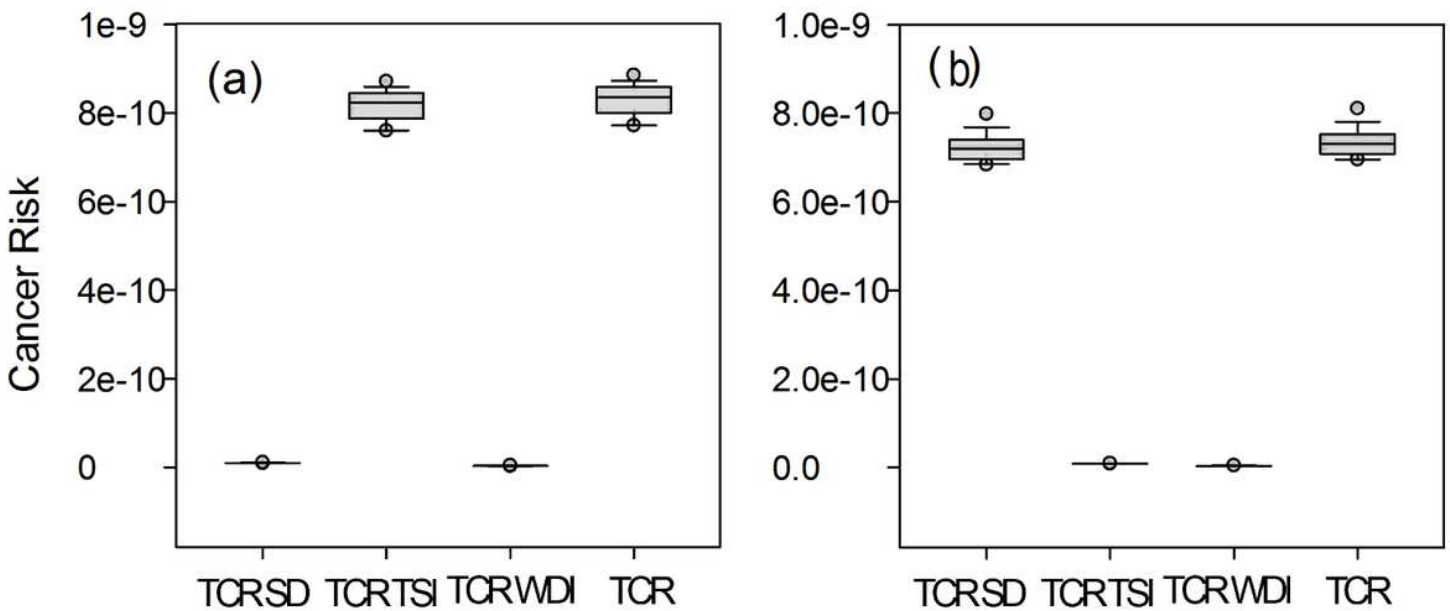


Figure 3

Cancer Risk through Soil and Paddy Water in Guangxi and Hunan Note: a:Hunan (adult); b):Guangxi (adult); TCRSD: cancer risk of the dermal contact through soil ; TCRTSI: cancer risk of the ingestion

through soil; TCRWD: cancer risk of the dermal contact through soil; TCR: sum of the cancer risk through soil and water through dermal and ingestion

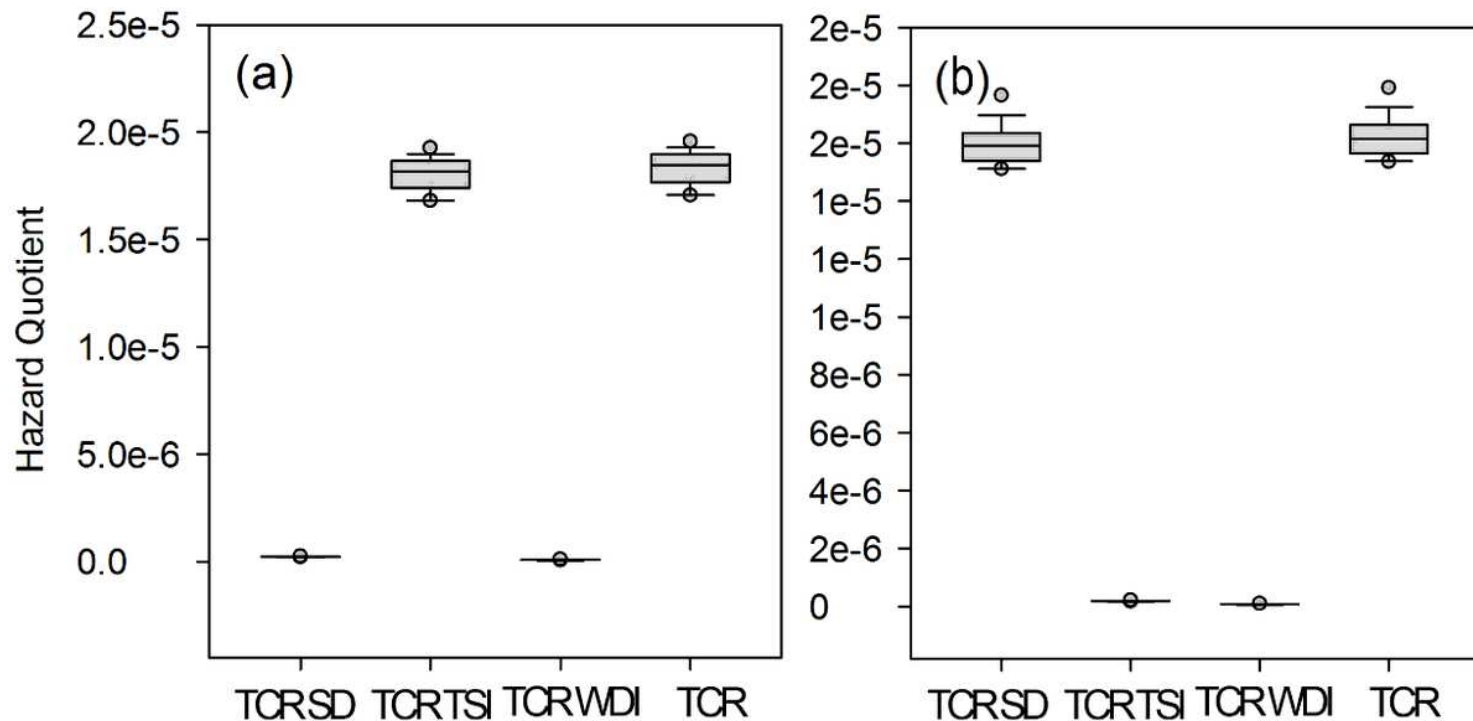
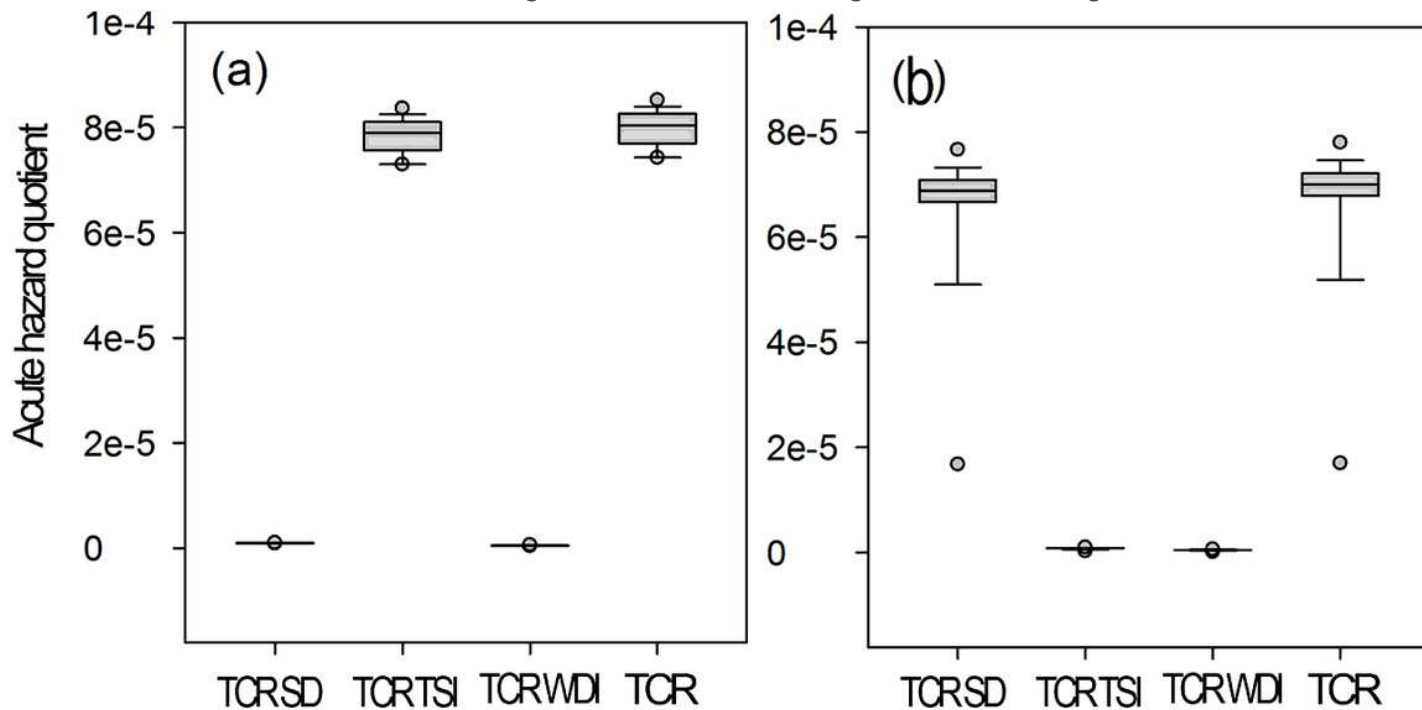


Figure 4

Non-Carcinogenic Risk for Lifetime Dose through Soil and Paddy Water in Guangxi and Hunan Note: a:Hunan (adult); b):Guangxi (adult); TCRSD: non-cancer risk of the dermal contact through soil ; TCRTSI: non-cancer risk of the ingestion through soil; TCRWD: non-cancer risk of the dermal contact through soil; TCR: sum of the non-cancer risk through soil and water through dermal and ingestion



## Figure 5

Non-Carcinogenic Risk Quotient from Acute Dose through Soil and Paddy Water in Guangxi and Hunan

Note: a:Hunan (adult); b:):Guangxi (adult); TCRSD: non-cancer risk of the dermal contact through soil ; TCRTSI: non-cancer risk of the ingestion through soil; TCRWD: non-cancer risk of the dermal contact through soil; TCR: sum of the non-cancer risk through soil and water through dermal and ingestion

## Supplementary Files

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