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Identification of key factors affecting neonicotinoid residues in crops and risk of dietary exposure☆

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ABSTRACT

Neonicotinoids, widely used on farmland, are ubiquitous in food; however, their distribution among various crops and associated exposure risks at the provincial level in China remain unclear. We collected 19 types of crop samples (fruits, vegetables, and tea) from farmland in nine prefectural cities in Zhejiang Province, China. We analyzed nine commonly used neonicotinoids in the edible portions of these crops. A notable detection rate (42.1 %–82.9 %) and high residual neonicotinoid concentrations (278 ± 357 ng/g) were observed. Tea exhibited the highest residue, followed by fruits, and vegetables showed the lowest ($P < 0.05$). Neonicotinoid ratios in crops to soil (R_C/S) and soil to water (R_S/W) were defined to discern insecticide distribution across different environments. Increased water solubility leads to increased migration of neonicotinoids (R_S/W) from agricultural soils to water through runoff, thereby increasing the relative contribution of nitenpyram and dinotefuran in water. In comparison with other studied compounds, all crops demonstrated the strongest soil uptake of thiamethoxam, denoted by the highest R_C/S value. Elevated R_C/S values in tea, pickled cabbage, and celery suggest increased susceptibility of these crops to neonicotinoid absorption from the soil ($P < 0.05$). Estimated dietary intake for teenagers, adults and elders was 8.9 ± 0.5, 8.9 ± 0.6, and 8.8 ± 0.3 μg/kg/d, respectively, below the reference dose (57 μg/kg/d). Teenagers, compared to adults and elders, exhibited significantly higher neonicotinoid exposure through fruit consumption, emphasizing the need for increased attention to neonicotinoid exposure among vulnerable populations.

1. Introduction

Neonicotinoid insecticides, known for their high efficacy, broad spectrum, and minimal toxicity to non-target organisms, have found extensive application in agricultural production (Lu et al., 2018; Sánchez-Bayo, 2018). Neonicotinoids are employed for crop pest management using diverse methods such as foliar sprays, seed treatment, and soil drenches (Bonmatin et al., 2014). This application leads to high residual concentrations of neonicotinoids in various environmental media, including agricultural soils, surface water, and especially in various crops and foods (Chang et al., 2018; Chen et al., 2022; Li et al., 2020a; Lu et al., 2018; Mahai et al., 2021; Zhang et al., 2019). Previous studies have demonstrated the widespread presence of neonicotinoids in foodstuff (Chen et al., 2020; Farajzadeh et al., 2017; Lu et al., 2018), honey (Kavanagh et al., 2021; Song et al., 2018; Wang et al., 2020), drinking water (Klarich et al., 2017; Li et al., 2020a), and other environments (i.e., air, indoor dust, soil, and surface water) (Forero et al., 2017; Mahai et al., 2021; Starner and Goh, 2012; Wang et al., 2019).

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Due to the inability to entirely remove neonicotinoids from fruits or vegetables through peeling or cleaning processes, dietary ingestion may present potential health risks to humans (Zeng et al., 2013; Zhang et al., 2019). For example, Zhang et al. (2019) reported detection of at least one neonicotinoid compound in a variety of foods at an elementary school in Hangzhou, China, with high concentrations of neonicotinoid residues (Zhang et al., 2019). Chen et al. (2014) detected neonicotinoids in fruits, vegetables, and honey from a grocery store in Boston, the United States, with the highest detection rate of imidacloprid and at least 90 % of honey containing neonicotinoid insecticides (Chen et al., 2014). Concentrations of neonicotinoids in fruit juices in Guangdong Province of China ranged from 0.07 to 3.93 ng/mL (Li et al., 2020a).

Recent extensive investigations have been conducted on neonicotinoid residues and their associations with health outcomes in humans (Cimino et al., 2017; Han et al., 2018; Taira et al., 2021; Zhang et al., 2018). Numerous reports have also documented neonicotinoid residue levels in serum, blood, and urine samples from various regions, including China, Japan, the United States, and other countries (Li and Kannan, 2020; Song et al., 2021; Ueyama et al., 2015; Xu et al., 2021; Zhang et al., 2022a; Zhang et al., 2022b). All these investigations indicated that the general population experiences widespread exposure to neonicotinoids, with dietary intake being a primary route of exposure. Hence, to estimate human exposure through the neonicotinoid-rich diet, residues must be detected in commonly consumed foods or crops (such as vegetables, fruits, wheat, rice, etc.).

Only about 5 % of the active neonicotinoids are directly absorbed by various crops, primarily found in farmland, including soil and surface water (Wood and Goulson, 2017). Less than 28 % of neonicotinoid insecticides are absorbed by the roots of crops, most of which remain in soil through seed treatment (Hu et al., 2023; Schaeffer and Wijntjes, 2022). Neonicotinoids in agricultural soil exhibit diverse behaviors, including adsorption, degradation, desorption, and crop migration, collectively influencing the residual levels and distribution of neonicotinoids in various environmental media (Mahai et al., 2021; Starner and Goh, 2012). The pronounced water solubility of neonicotinoids is identified as a pivotal factor influencing their leaching, runoff, and transport from soil to surface water (Burton et al., 2004; Martin et al., 2012). The findings indicate a sequential increase in the leaching capacity of thiamethoxam, imidacloprid, and dinotefuran, significantly correlated with rising aqueous solubility (Kurwadkar et al., 2014). Dinotefuran (DIN), characterized by high water solubility, is commonly regarded as a predominant neonicotinoid compound in surface water (Chen et al., 2019a; Xiong et al., 2021). However, to date, limited studies have investigated the soil-to-crop and soil-to-water transport of neonicotinoids, along with their influential factors and mechanisms.

Therefore, in this field study, the neonicotinoid residues were quantified in various crop types, including fruits, vegetables, and tea, within Zhejiang Province, China. This study aimed to investigate the distribution patterns of various neonicotinoids between soil and crops, and between soil and water, in relation to the neonicotinoid residual concentrations found in corresponding soil and surface water samples simultaneously collected from the same agricultural lands within the study area. Additionally, the investigation sought to identify the primary influential factor governing the migration of neonicotinoids across different environmental media. Furthermore, an assessment of the general population's exposure to neonicotinoids through overall dietary intake has been conducted, providing a foundational perspective for a comprehensive understanding of potential health risks associated with neonicotinoid contamination in agricultural lands.

2. Methods and Materials

2.1. Sampling campaign

In 2021, samples of vegetables (including amaranth, bok choy, broad beans, celery, celtuce, Chinese cabbage, cilantro, garlic, lettuce, mustard, peas, pickled cabbage, pickled mustard tuber, rape, spinach, spring onion, white radish), fruits (specifically oranges), and tea, were randomly collected from 76 farmlands located across nine prefectural cities in Zhejiang Province, with the exclusion of Huzhou and Zhoushan islands. A total of 85 representative samples of crops with similar growth stage (all approaching the stage of maturation ready for harvest) were collected in two or more different locations in each municipality. Therefore, the representative crop samples here can to some extent reflect the real dietary exposure situation of the studied population. It is worth noting that in this study, we exclusively collected oranges as the sole fruit category. According to prior literature, variations in neonicotinoid insecticides among different fruits within the same research area are minimal, unlike the substantial differences observed in vegetables and tea (Lu et al., 2018). Additionally, oranges rank among the most prevalent and highest-selling fruits in Zhejiang Province. Therefore, collecting oranges exclusively here serves as a reasonably representative dietary exposure for most fruits, avoiding significant bias. In addition, soil samples and adjacent surface water samples were simultaneously collected at the same sampling points (farmlands) of the crop samples. The detailed sampling procedures for soil and water samples have been detailed described in our previous studies (Chen et al., 2024; Chen et al., 2022). Concisely, for each sampling site (cropland), we collected five soil samples horizontally and randomly at a depth of approximately 15 cm from the surface, and these samples were then blended to form a representative composite. The surface water samples were obtained by combining three samples collected at depths of 50, 100, and 150 cm from the water surface, employing a Kemmerer-type water sampler to collect approximately 1.0 L of water. Detailed sampling sites (farmlands) and sample sizes by prefecture-level cities and crop types are provided in Fig. 1 and Table S1 of the Supporting Information. All the samples (crops, soil, and water) were stored at −20 °C refrigerator before laboratory analysis.

2.2. Reagents and materials

Nine neonicotinoid standards, including acetamiprid (ACE), thiamethoxam (THIA), thiacloprid (THI), imidacloprid (IMI), clothianidin (CLO), dinotefuran (DIN), nitenpyram (NIT), imidaclothiz (IMID), and flonicamid (FLO), were purchased from Dr. Ehrenstorfer (Augsburg, Germany). The internal/isotopically labeled standards, including IMI-*d4*, THIA-*d3*, and CLO-*d3* were purchased from C/D/N Isotopes Inc (Quebec, Canada). QuEChERS kits were obtained from the Shimadzu Corporation (Kyoto, Japan), and acetonitrile of HPLC grade was purchased from Fisher Scientific (Leicestershire, UK).

2.3. Extraction and cleanup

Neonicotinoid analysis in crops and soil samples was performed using the QuEChERS method. Detailed information on the pretreatment of soil samples can be found in our earlier publication (Chen et al., 2022). For the crop samples, fruit and vegetable samples were washed with tap water and wiped down with paper towels, then ground to a paste and weighed in a 50 mL centrifuge tube at 10 g (2 g of tea). Next, 10 μL of recovery surrogates (IMI-*d4*, THIA-*d3*, 10 μg/mL) and 10 mL of acetonitrile were added to the tube and then treated with a packet of QuEChERS salt (containing 4 g of $MgSO_4$, 1 g of NaCl, 500 mg of disodium citrate). The tube was then vortexed for 2 min and then centrifuged at 4000 rpm for 5 min. The supernatant was transferred to a SPE column containing 25 mg of PSA, 7.5 mg of GCB, and 150 mg of $MgSO_4$. After vortex and centrifugation, the supernatant was loaded into a 2 mL centrifuge tube, blow-dried with nitrogen and reconstituted with 200 μL of 15 % acetonitrile/water. The internal standard of 100 ng (clothiandin-d3) was added to the centrifuge tube and then transferred through a 0.22 μm membrane into the sample vial for instrument analysis. For the water samples, the liquid-liquid extraction method was employed according to our previous study (Chen et al., 2024).

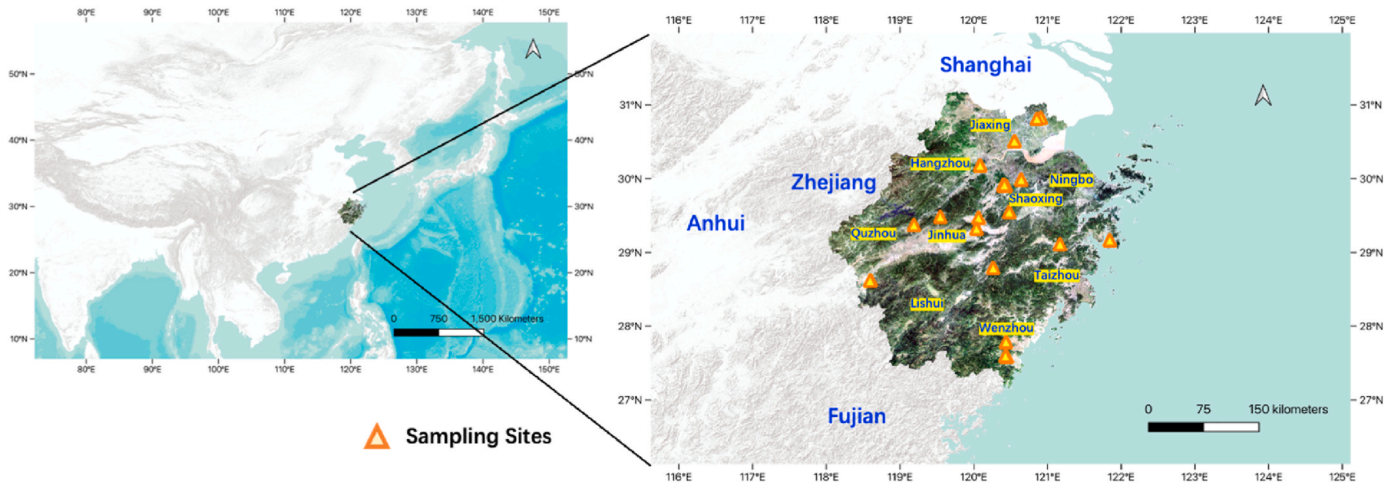

Fig. 1. The geographical illustration of the sampling sites in Zhejiang Province, China.

2.4. Instrumental analysis and QA/QC

Quantitative conditions refer to our previous studies (Chen et al., 2023; Chen et al., 2019b). The nine individual neonicotinoids were identified using the ultra-performance liquid chromatography coupled with a triple quadrupole mass spectrometer Xevo TQ-s (UPLC-MS/MS) (Waters Corporation, Milford, MA, USA). Chromatographic separation was performed on an Acquity HSS T3 column (50 mm × 3.0 mm, 1.8 μm, Waters Corporation), which was maintained at 35 °C. A 5 μL aliquot of the extract was injected and the mobile phase flow rate was set at 0.2 mL/min. The mobile phase consisted of 0.1 % formic acid in ultrapure water (A) and methanol (B) using gradient elution (Table S2). Target analytes was analyzed by positive electrospray ionization and multiple reaction monitoring modes.

Blank samples were injected into each of the 8 samples to detect contaminants in the reagents and materials. Matrix analysis was performed by injecting 0.5, 5, and 50 ng/mL of the target analyte into randomly selected crop samples, and recovery rates for neonicotinoids ranged from 88 % to 107 %. Ten and three times of signal-to-noise ratio (SNR) of qualitive ion was set as the limit of quantification (LOQ) and limit of detection (LOD). The LODs and LOQs for target analytes were 0.01 ng/g to 0.03 ng/g and 0.03–0.08 ng/g, respectively (detailed information see in Table S3). In addition, the detailed QA/QC for the determination of neonicotinoids in corresponding soil and water samples can be referred to the two previous studies, respectively (Chen et al., 2024; Chen et al., 2022).

2.5. Data analysis

The relative potency factor (RPF) method was used to assess the overall dietary risk of all neonicotinoids, we integrated different individual neonicotinoids into a single metric (IMI_{RPF}) using the toxicity value of IMI as a reference. The RPF values for different neonicotinoid compounds are shown in Table S4. However, the reference dose (RfD) for IMID, NIT and FLO was not available in current study, so the same RfD for IMI was used in the calculation:

$$IMI_{RPF}(ng/g) = \sum neonicotinoid_i(ng/g) \times RPF_i = IMI + THIA \times 9.5 + ACE \times 0.803 + CLO \times 5.816 + THI \times 14.25 + DIN \times 2.85 + IMID + NIT + FLO$$

where *i* represents the different neonicotinoids.

The estimated dietary intake (EDI) of total neonicotinoids for Zhejiang population was estimated according to the handbook of Chinese Population Exposure Parameters (Duan, 2013; Yang et al., 2018), which was calculated using the following equation:

$$EDI(\mu g/kg/d) = C_i(ng/g) \times UI_i(g/day) \times 10^{-3}(\mu g/ng)/BW(kg)$$

where C_i represents the concentration of each neonicotinoid or IMI_{RPF} detected in the crop samples, UI_i represents the average food consumption data for each crop from Zhejiang Province (Table S5). The mean level of body weights (BW) was 45 and 63 kg for age groups of teenagers and adults/elders, respectively. The uncertainty of EDI estimation in different age groups was quantitatively calculated by Monte Carlo Simulation method based on the distribution of IMI_{RPF} with mean and standard deviation. And 100,000 times simulation run for each age groups.

The concentration ratios of neonicotinoids in crops to soil and water to soil were defined as R_C/S and R_W/S to evaluate the distribution and transport of neonicotinoid insecticides in different environmental media (which are all detailed discussed in the **3.4 Section**). In addition, the high spatial resolution distribution of neonicotinoids in surface water and soil in Zhejiang Province was obtained by inverse distance weighted interpolation (Fig. S1 and Fig. S2).

All statistical analyses were performed using SPSS 22.0 software (Statistical Product and Service Solutions, IBM, USA) and R studio software (version 2022.12.0 + 353, POSIT Corporation, MA, USA). One-way ANOVA was used to compare the difference of neonicotinoid residues in different types of crops. The relationship between neonicotinoid residual concentration and other environmental parameters (Log*Kow*, water solubility) was investigated by Pearson correlation test. The statistically significant level was $P < 0.05$.

3. Results and discussion

3.1. Neonicotinoid residues in different crops and composition profiles for individual compounds

Neonicotinoids were extensively identified in various crops, with detection rates ranging from 42.1 % to 82.9 %. Each crop exhibited a minimum of three individual neonicotinoid compounds, with THI displaying the highest detection rate at 82.9 %, followed by IMI (77.6 %), NIT (64.5 %), and THIA (60.5 %) (Table S3). The average residual concentration with standard deviation of total neonicotinoids ($\sum$NEOs) was 278 ± 357 ng/g and the coefficient of variation was 1.28. The high variation was attributed to the different sampled sites and types of crops (discussed below). The studied crops exhibited relatively higher average levels of THIA (156 ± 220 ng/g), CLO (77.3 ± 181 ng/g), IMI (64.7 ± 146 ng/g), and NIT (59.7 ± 103 ng/g) compared to other compounds. In comparison with the average $\sum$NEOs in crops across nine prefectural cities (Table S6), Hangzhou had the highest statistically significant $\sum$NEOs level at 589 ± 424 ng/g ($P < 0.05$), followed by Ningbo and Shaoxing, with levels of 305 ± 208 and 249 ± 428 ng/g, respectively. The significantly lowest residue levels were found in Taizhou (65.0 ± 46.0 ng/g) and Lishui (83.1 ± 56.4 ng/g) ($P < 0.05$). $\sum$NEOs in crops exhibited a statistically higher level in Hangzhou than in Lishui ($P = 0.016$), Quzhou ($P = 0.042$) and Taizhou ($P = 0.013$). While pesticide application practices exhibit minimal variation across cities, spatial disparities in neonicotinoid residual concentrations may be attributed to the diverse crop types collected in different cities (Whalen et al., 2021).

The residues of the nine neonicotinoids in vegetables, fruits (specifically oranges) and tea were 329 ± 332, 777 ± 209 and 1567 ± 341 ng/g, respectively. The neonicotinoid residue concentration in tea was significantly higher ($P < 0.05$) due to increased insecticide use in tea planting. This observation aligns with our prior study, indicating higher neonicotinoid residues in the soil of tea-planted agricultural land compared to other crops (Chen et al., 2022). Additionally, Xiao et al. (2023) observed elevated levels of total target neonicotinoids in tea samples representing the Jiangnan area of China. Within various vegetable species, the significantly highest concentrations of neonicotinoids were found in pickled cabbage, celery, and mustard with levels of 1171 ± 294, 656 ± 101, and 645 ± 170 ng/g, respectively. The cultivation of these three crops involves two methods, namely, seed dressing and pesticide spraying, potentially leading to relatively higher levels of insecticide residues in these crops. Fruits, such as oranges, also exhibited high concentrations of neonicotinoids at 777 ± 209 ng/g (Fig. 2a).

Regarding the composition profiles of neonicotinoid residues, THIA was found to be the main compound in majority of crops, contributing from 0 % to 82.6 %, notably in tea (67.5 %), peas (52.4 %), and oranges (82.6 %), while undetected in spinach, cilantro, spring onion, and pickled cabbage (Fig. 2b). Furthermore, NIT was extensively detected in various crops, exhibiting high concentrations of 897 (contribution:76.6 %), 252 (38.3 %), and 201 ng/g (12.8 %) in pickled cabbage, celery, and tea, respectively. High concentrations of THIA and NIT in tea were consistent with previously detected high mean concentrations (650 (THIA) and 54 ng/g (NIT)) of neonicotinoids in Japanese green tea (Nimako et al., 2021). Higher CLO concentration in bok choy (426 ng/g) and spring onion (275 ng/g) was also the main neonicotinoid for both crops, accounting for 71.4 % and 66.1 %, respectively. The high levels of

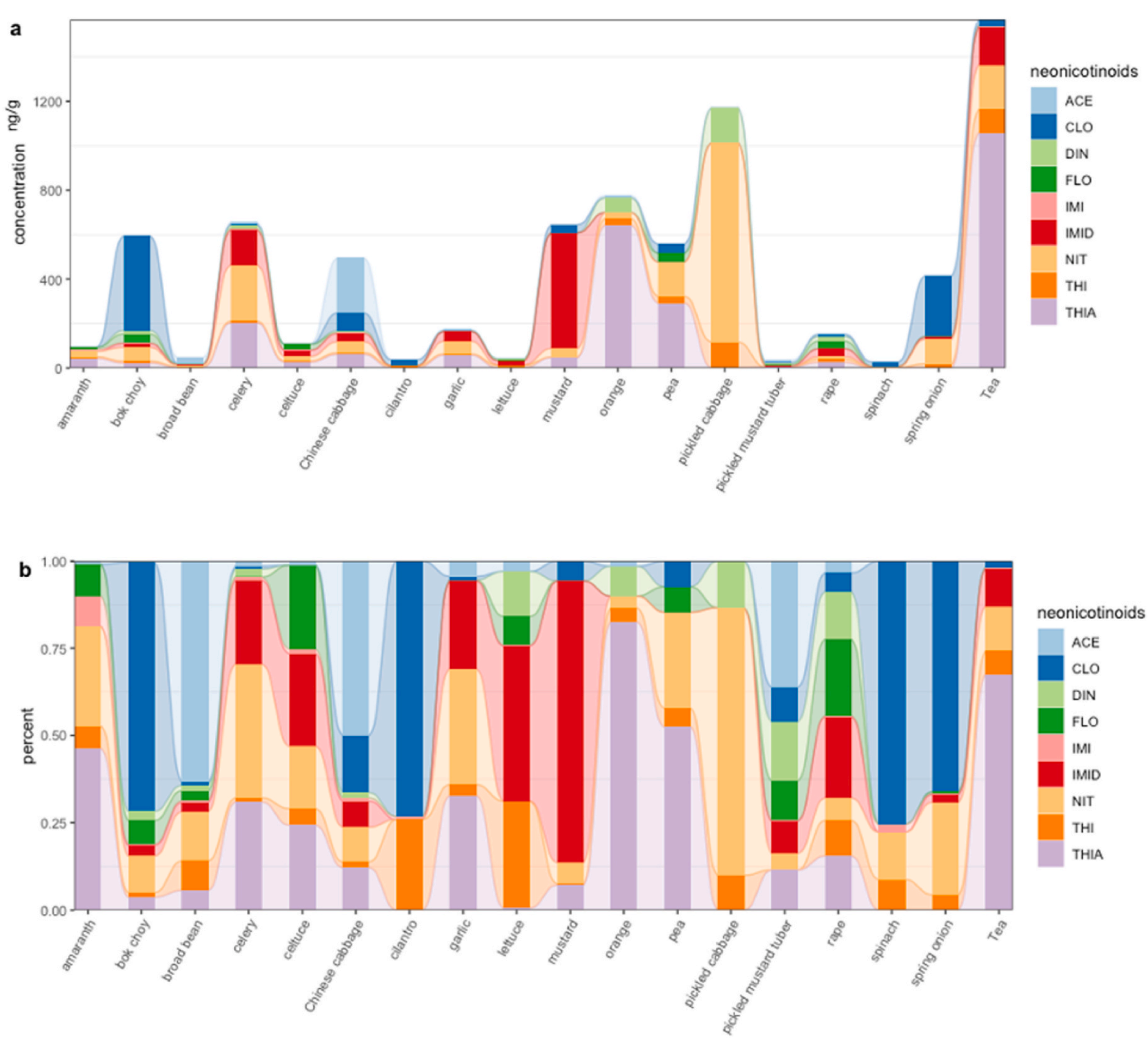

Fig. 2. Residual concentrations of nine individual neonicotinoids in different crops, including vegetables, fruits (oranges), and tea (**a**) and the relative contribution (percent) of different neonicotinoid compounds in different crops (**b**).

THIA and CLO in crops, particularly bok choy and spring onion, may result from their frequent application via seed coating. THIA and CLO are also widely used as seed coating treatments in the United States, making them the most widely detected class insecticides in the U.S. history (Simon-Delso et al., 2015).

3.2. Comparison with previous studies in different crops and regions

In this study, the concentrations of individual neonicotinoids in fruits, vegetables, and tea were compared with previous studies conducted across different regions and countries globally (Table S7). Previously, Lu et al. (2018) assessed neonicotinoid levels in fruits and vegetables commonly consumed in a cafeteria in the United States (USCC) and a primary school in Hangzhou, China (Lu et al., 2018). It was found that the neonicotinoid concentrations in fruits and vegetables in Hangzhou were comparable between *Lu* et al.'s results (112 ng/g) and our study (295 ng/g), and both were relatively higher than that in the cafeteria the United State (21.2 ng/g) (Lu et al., 2018). Furthermore, the levels of neonicotinoid residues identified in fruits and vegetables gathered from Boston (USA) markets (150 ng/g) were comparatively lower than those in this study (Chen et al., 2014). These findings suggest that neonicotinoid contamination in agricultural products in China surpasses that observed in the United States.

Tan et al. (2016) explored the presence of various neonicotinoids in vegetables (21.6 ng/g) and fruits (4.77 ng/g) in the Beijing market. They identified NIT (7.83 ng/g in vegetables, not detected in fruits) and IMI (1.55 vs 1.07 ng/g) as the predominant neonicotinoids in these samples. However, it's noteworthy that the residue levels in their findings were comparatively lower than those observed in the current study (Tan et al., 2016). In the 5th and 6th Chinese Total Diet Studies, the individual neonicotinoid - ACE, IMI, CLO, and THIA – exhibited higher detection rates (ranging from 20 % to 100 %) and relative contributions in fruit and vegetable samples, consistent with our findings (Chen et al., 2020). The residual concentration of neonicotinoids in fruit and vegetable samples in Beijing was lower than that in Zhejiang Province (mean: 31.03 ng/g *vs* 188.41 ng/g), which also verify that pollution of neonicotinoids in crops in Zhejiang Province was more severe compared with that in the northern China. Moreover, *Chen* et al. *(2020)* observed a significant increase in the concentration of THIA and CLO in vegetables during the 6th Chinese Total Diet Studies compared to the 5th Chinese Total Diet Studies (Chen et al., 2020). This suggests a notable rise in the usage of THIA and CLO in agriculture in recent years, potentially explaining the elevated concentration and contribution of THIA and CLO in most crops in our study. Overall, various factors, such as neonicotinoid levels in agricultural soils and surface waters (Xiao et al., 2023), pesticide usage patterns, agricultural practices, and others, may contribute to variations in neonicotinoid residues across different geographical areas and crop types.

3.3. Dietary exposure risks of neonicotinoids for Chinese general population

While the residual concentration of neonicotinoids in all crops samples (ranging from 0.15 to 951 ng/g) in Zhejiang Province did not exceed the national standards for food safety in China (10 mg/g) (GB2763-2014) (Beijing: Ministry of Agriculture of the People's Republic of China, 2014) and the United States Federal Regulations (50 mg/g) (40 CFR Part 180-Tolerances and exemptions for pesticide chemical residues in food) (U. S. Environmental Protection Agency, 2002) (except for the residual concentrations of THIA in legume crops (59.0 ng/g *vs* 20 ng/g), which may be due to differences in dietary habits and pesticide application in different countries). Previous studies have consistently highlighted dietary intake as the primary exposure pathway to neonicotinoids for the general population, contributing to more than 80 % through diet (Zhang et al., 2023; Zhang et al., 2019). Therefore, considering the potential health risks associated with ingestion, neonicotinoid residues in crops cannot be disregarded.

In this study, the IMI_{RPF} values varied among different crops, ranging from 74.1 ± 11.6 ng/g (white radish) to 4361 ± 2360 ng/g (tea). Tea, fruits (specifically oranges), and vegetables exhibited IMI_{RPF} values of 4361 ± 2360, 1764 ± 1518, and 805 ± 1413 ng/g, respectively. Statistical analysis revealed that tea had the highest IMI_{RPF} value, followed by fruits (specifically oranges), with vegetables showing the lowest levels ($P < 0.05$) (Fig. 3a). This trend aligns with findings from a prior study that investigated neonicotinoid levels in tea and various crops (Li et al., 2020b). EDIs of neonicotinoids were calculated for light-colored vegetables, dark-colored vegetables, fruits (specifically oranges), and tea among teenagers, adults, and elders, considering their daily consumption habits (seen in the **Methods and Materials** section) (Fig. 3b).

The mean 95 % cumulative EDIs were 8.9, 8.9, and 8.8 μg/kg/d for teenagers, adults, and elders, with corresponding 95 % confidence of intervals (CIs) of 6.0–11.9, 5.9–11.8, and 5.9–11.8 μg/kg/d. There was non-significant difference between crops at different age groups ($P >$ 0.05). Despite the relatively higher neonicotinoid residues in tea, the EDIs from tea exposure were comparatively lower than those from vegetables and fruits, attributed to its lower consumption. Light-colored

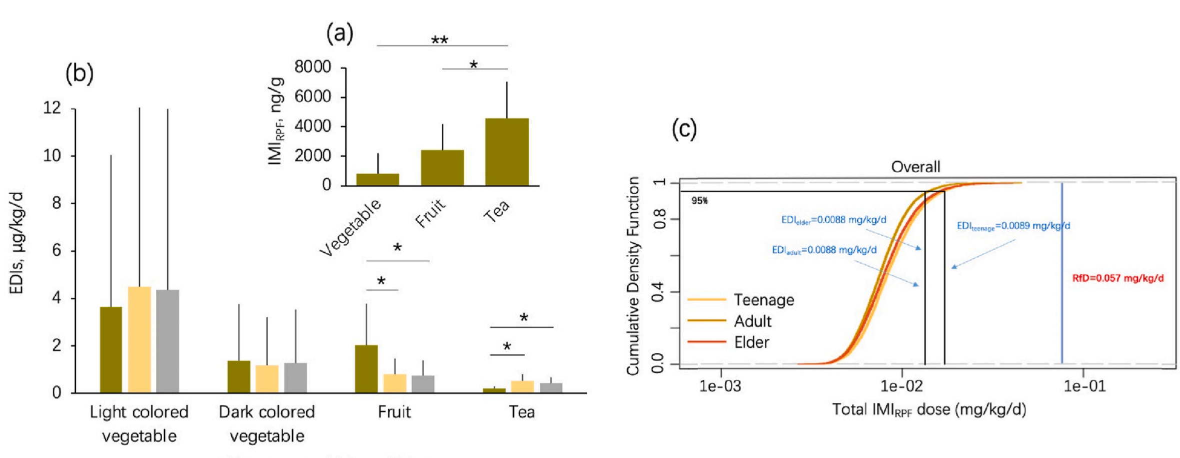

Fig. 3. The IMI_{RPF} values of different crop types, including vegetables, fruits (oranges) and tea (*, $P < 0.05$; **, $P < 0.01$) (**a**) and EDI of different crops (the vegetables are divided into light colored and dark colored species) (**b**). The cumulative distributions of the estimated dietary intakes (EDIs) of total neonicotinoids from fruits and vegetables consumption by teenagers, adults, and elders age groups (**c**). EDIs of teenagers, adults and elders corresponding to 95 % cumulative percentage are also shown.

vegetable exhibited the highest EDIs across different age groups, indicating greater application amounts of neonicotinoids for peat control. For fruits, the teenagers exposed to significant higher level of neonicotinoids (mean: 5.3 μg/kg/d, 95 % CI: 3.4–7.2 μg/kg/d) compared to those of adults (2.1 μg/kg/d, 95 % CI: 1.3–2.8 μg/kg/d) and elders (1.9 μg/kg/d, 95 % CI: 1.3–2.6 μg/kg/d) ($P < 0.05$), which resulted in higher potential exposure risks. In addition, in tea, the EDI for teenagers (0.083 μg/kg/d, 95 % CI: 0.0022–0.16 μg/kg/d) was significantly lower than that for adults (0.21 μg/kg/d, 95 % CI: 0.0056–0.41 mg/kg/d) and elders (0.18 μg/kg/d, 95 % CI: 0.0048–0.36 μg/kg/d) also due to the significantly fewer daily consumption. It is recommended to pay more attention to the fact that children are exposed to more neonicotinoids after consuming fruit. Furthermore, Fig. 3c illustrates the cumulative distribution of neonicotinoid EDI for the general population based on varying consumption levels of vegetables, fruits, and tea in China. The 95 % cumulative EDIs in this study remained consistently below the RfD threshold of 57 μg/kg/d for chronic health effects. In summary, the neonicotinoid exposure levels within the general population in China are deemed acceptable. However, attention needs to be paid to the long-term exposure to some vulnerable population.

3.4. Distributing difference of various neonicotinoids in different crops and environments

In Fig. 4a, the composition profiles of individual neonicotinoids in crops, soil, and surface water (rivers near corresponding sampling sites of crops and agricultural soils) are depicted. We postulate a uniform pesticide application method for the majority of crops. As a result, the comparative analysis of the extensive distribution of various neonicotinoids in soil, water, and crops within the same agricultural field provides insights into the migration processes and potential mechanisms of these insecticides across different environmental media. Evidently, IMI constituted the predominant neonicotinoid compound in the agricultural soil, representing 57.9 % of the total, while the proportions of NIT and DIN in the soil were only 4.2 % and 7.2 %, respectively. In contrast, NIT and DIN emerged as the primary neonicotinoids in surface water (rivers near the agricultural lands) with relative contributions of 22.8 % and 31.1 %, respectively, surpassing their proportions in agricultural soil. The relative contribution of IMI in surface water (21.2 %) was notably lower than that in agricultural soil. Residual neonicotinoids in farmland permeate nearby water bodies, specifically rivers, through runoff. Consequently, NIT and DIN, characterized by high water solubility, are predisposed to enter these water bodies, leading to elevated concentrations and contributions compared to other neonicotinoid compounds. This observation aligns with the findings of our earlier study, which identified NIT and DIN as the primary neonicotinoids in surface water/rivers in the Yangtze River Delta region (Chen et al., 2019a). In addition, Fig. 4b shows that Log*Kow* is significantly negatively correlated with log-transformed R_W/S (concentration ratio of neonicotinoids in water to soil) (LogR_W/S) ($P = 0.032$), suggesting that more water-soluble substances are more likely to enter the rivers in nearby agricultural lands.

THIA and IMI emerged as the predominant neonicotinoids in various crops, constituting 25.5 % and 21.2 % of the total, respectively. The concentration ratio of neonicotinoids in crops to soil (R_C/S) for THIA, NIT, FLO, DIN, THI, IMID, and CLO all exceeded 1, while R_C/S for IMI and ACE fell below 1. Among the various individual neonicotinoids, THIA exhibited the highest R_C/S value (at 135), surpassing other compounds. This indicates that crops possess a substantial capacity for the uptake of THIA from agricultural soil, a phenomenon supported by previous field evidence (Li et al., 2018). It is known that organic pollutants in soil are typically transported from the root system to the ground through the transpiration process by plants. Consequently, neonicotinoids with high water solubility are expected to be more easily absorbed by crops from farmland soil. Fig. 4c illustrated the negative correlation between Log-transformed R_C/S and Log*Kow*, however, a non-significant correlation was observed between them ($P = 0.258$). This suggests that the distribution of neonicotinoids between crops and soil is not solely controlled by the uptake and transpiration processes of different crops, as some crops receive neonicotinoids through both spraying and seed coating.

Moreover, Fig. 5 and Table S8 present the distribution variations of individual neonicotinoids between crops and soils across different croplands. Different crops addressed considerable difference in uptake capacity of neonicotinoids from agricultural soils. Tea, pickled cabbage, and celery exhibited comparatively higher R_C/S levels, whereas pickled mustard tuber and broad bean displayed lower R_C/S levels. Substantial variations were observed in the relative contribution of R_C/S (R_C/S percent) for different neonicotinoid compounds (see Fig. 5a). The R_C/S percent of IMI varied among different crops, ranging from 0 % (mustard, pea, and pickled cabbage) to 8.7 % (amaranth), which was notably lower than that of other compounds. This observation suggests that IMI

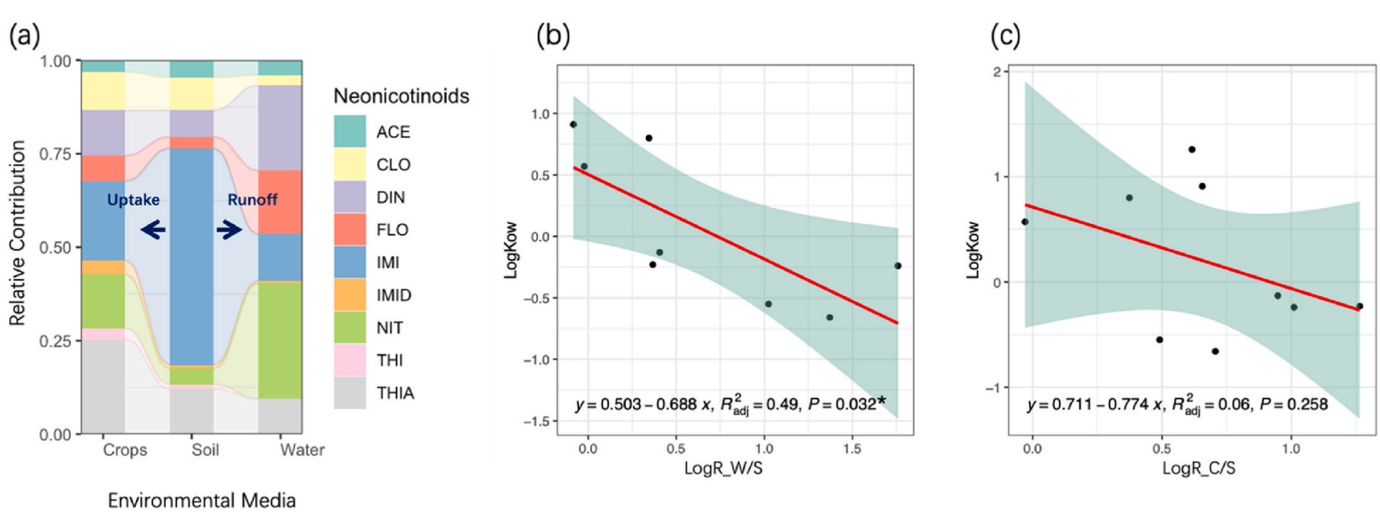

Fig. 4. The composition profiles of individual neonicotinoids and their distribution in different environmental media influenced by Log*Kow* value. The relative contributions of nine neonicotinoids in crops, soil, and water (**a**). The correlation between log-transformed concentration ratio of water to soil (LogR_W/S) and value of Log*Kow* for different compounds (**b**). The correlation between log-transformed concentration ratio of crops to soil (LogR_C/S) and value of Log*Kow* for different compounds (**c**). The regression model, adjusted *R* square (R^2_{adj}), *P*-value, and 95 % confidence of intervals (95 % CIs) are all shown. * represents the statistically significant level ($P < 0.05$).

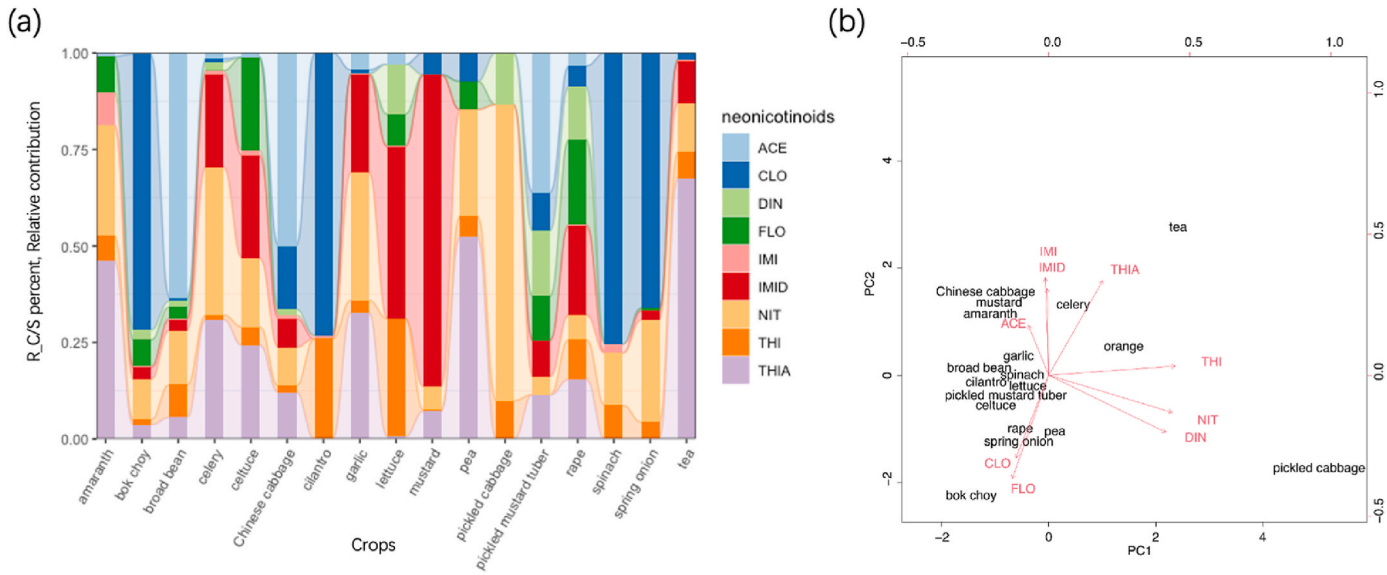

Fig. 5. The residual concentration ratios of nine individual neonicotinoids in different crops to corresponding agricultural soil (R_C/S), including vegetables, fruits (oranges), and tea. (a) The relative contribution of the neonicotinoid ratios of crops to soils (R_C/S percent). (b) The principal component analysis (PCA) (PC1 and PC2) identifies the difference of the ratios (R_C/S percent) for different crops.

tends to be distributed more in agricultural soils than in crops. The R_C/S percent for THIA, THI, NIT, and FLO was significantly higher ($P < 0.05$) in tea compared to various vegetables and fruit. This difference can be attributed to the predominant use of neonicotinoid spraying in tea cultivation, resulting in higher concentrations in tea than in agricultural soils. This is in contrast to the common practices of using seed coatings and soil drenches in vegetable and fruit cultivation. Moreover, irrigated tea cultivation involves higher water usage, leading to increased runoff and vertical infiltration of neonicotinoids into surface and groundwater (Chen et al., 2022). This process reduces residues in surface soil, contributing to the higher R_C/S percentage observed in tea.

Additionally, Fig. 5b delineates clusters representing the R_C/S percentages of various compounds across distinct crops. Evidently, there is a discernible inclination for NIT and DIN to be distributed on pickled cabbage. Based on the above-discussed results, it can be inferred that pickled cabbage primarily absorbs NIT and DIN through transpiration due to their relatively high water solubility. Tea, on the other hand, accumulates a significant amount of THIA, suggesting that THIA is predominantly applied as a spray in tea cultivation, and tea plants efficiently absorb THIA from agricultural soil. In contrast, crops such as rape, spring onion, and peas exhibit lower THIA concentrations, indicating limited THIA spraying. Instead, these crops primarily utilize seed dressings, involving CLO, FLO, NIT, and DIN, and absorb these neonicotinoids through transpiration. The different absorption capacity of different crops to different neonicotinoid insecticides will affect the residue distribution of neonicotinoids in the crops and soils. Within this study, we examine the distribution patterns of various neonicotinoids in crops, soil, and surface water in real-world agricultural systems. These observations serve as the basis for inferring their migration tendencies. However, the absence of well-controlled conditions, particularly concerning pesticide application and other environmental factors, hinders precise inference regarding the differential uptake capacities of different crops for neonicotinoids and their underlying mechanisms. Therefore, further analysis of the different uptake capacities of insecticides by crops through control experiments will help us to evaluate the distribution of neonicotinoids between crops and soils and identify the potential exposure risks to human health through diet.

4. Conclusions

In this field study, we analyzed nine individual neonicotinoids in various common crops across nine prefectural cities in Zhejiang Province, China. Our findings revealed significantly higher detection rates and residual concentrations of neonicotinoids in tea, fruits, and vegetables in Zhejiang Province compared to those observed in developed countries (e.g., the United States) and the northern part of China (e.g., Beijing). In assessing the residual concentrations of neonicotinoids in agricultural soil and surface water, we identified water solubility as the primary factor influencing the migration of water-soluble insecticides from soil to water. Additionally, our findings indicate that the absorption capacity of crops for insecticides from soil to crops is contingent upon physicochemical properties of different compounds, crop types, their respective application methods. Though the cumulative EDIs of the neonicotinoids for the residents, including teenagers, adults, and elders were all lower than the RfD value for chronic toxicity, the significantly highest exposure of neonicotinoids from fruits for teenagers age group should be paid more attention.

CRediT authorship contribution statement

Wenfei Yu: Writing – original draft, Methodology, Investigation, Data curation. **Ruxin Wu:** Methodology, Investigation, Data curation. **Li Zhang:** Investigation, Conceptualization. **Yangzhong Pan:** Methodology, Data curation. **Jun Ling:** Methodology, Data curation. **Dan Yang:** Methodology, Data curation. **Jiajia Qu:** Methodology. **Zhen Tao:** Methodology. **Ruirui Meng:** Methodology, Investigation. **Yuexing Shen:** Methodology, Investigation. **Jingtong Yu:** Methodology, Investigation. **Nan Lin:** Writing – original draft, Conceptualization. **Bin Wang:** Writing – original draft, Conceptualization. **Hangbiao Jin:** Writing – original draft, Conceptualization. **Meirong Zhao:** Writing – original draft, Conceptualization. **Yuanchen Chen:** Writing – review & editing, Writing – original draft, Visualization, Methodology, Conceptualization.

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 B. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2024.123489.

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