



Residual distribution and risk assessment of neonicotinoids in urban green space soils of the pearl river delta, South China: A socioeconomic analysis

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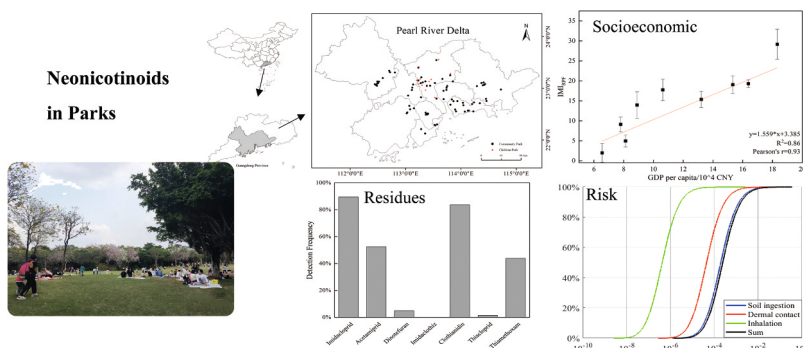
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HIGHLIGHTS

- First systematic NEOs soil study in parks.
- Assessed health risks of NEOs to humans.
- NEOs distribution compared in park soils.
- Economic growth linked to pollution.
- Nemerow index method applied to NEOs.

GRAPHICAL ABSTRACT



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ABSTRACT

Urban green spaces are the soil component in cities that interacts most closely with humans. This study investigated the residues of seven neonicotinoids (NEOs) in soils from urban green spaces within the Pearl River Delta (PRD) region and analyzed the correlation between the residue characteristics and the region's economic development. Notably, we introduced the Nemerow Index method, a comprehensive approach, to quantify the overall pollution level of NEOs in the soil of urban park green spaces and utilized this to assess the cumulative exposure probability risks for different populations in this scenario. We found that: (1) The soil of urban park green spaces exhibited varying degrees of NEOs contamination (Σ_7 NEOs: N.D.–137.31; 6.25 $\mu\text{g}/\text{kg}$), with imidacloprid and clothianidin constituting the highest proportions (89.46 % and 83.60 %); (2) The residual levels of NEOs in Children's Park were significantly higher than those in community parks within Guangzhou, with an average value of 13.30 $\mu\text{g}/\text{kg}$ compared to 3.30 $\mu\text{g}/\text{kg}$; (3) The residual characteristics of NEOs exhibited a positive correlation with regional economic development; specifically, the per capita GDP well correlated with IMI_{RPF}, a summation of seven NEOs in imidacloprid equivalents via relative potency factors ($R^2 = 0.86$). Regions with higher economic development typically exhibited elevated IMI_{RPF} levels; (4) The fitted cumulative probability distributions for average daily exposure doses revealed that children's exposure was an order of

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magnitude higher than adults'. Despite this, the exposure risks for both groups remained within acceptable limits.

1. Introduction

Neonicotinoids (NEOs) are a class of systemic neuroactive pesticides that primarily function by binding to the nicotinic acetylcholine receptors (nAChRs) on the post-synaptic membranes of insects [1,2]. Characterized by high efficacy, broad spectrum, and low toxicity to non-target organisms, they are widely used in pest control for crops such as rice, wheat, fruits, and vegetables [3,4]. However, only about 5 % of the active ingredients of NEOs are absorbed by the crops; the majority disperses into the environment [5,6]. In recent years, the non-agricultural uses of NEOs have rapidly expanded, combined with the indiscriminate prophylactic application patterns, resulting in frequent detections of NEOs in various environmental media worldwide, primarily including soil [7–9], water bodies, and sediments [10,11].

The negative impacts of NEOs on pollinators have been extensively studied, but these chemicals also present risks to aquatic life, mammals, and humans [12,13]. Research has demonstrated that NEOs can cause abnormal movement patterns in fish and disrupt reproductive processes in mice [14]. In humans, exposure can lead to symptoms such as nausea, vomiting, headaches, and diarrhea [15,16]. Urban green soil is one of the most accessible pathways for city dwellers to come into contact with pesticides. The residues of these chemicals in the soil can not only impede plant growth and biodiversity but also pose health risks to residents through various exposure routes, such as dust inhalation, respiratory uptake, and ingestion by children through hand-to-mouth activities. Despite these significant concerns, there is a notable lack of research on the extent of pesticide contamination in urban green spaces and the associated health risks.

This research focuses on the residual characteristics of NEOs in the soil of typical urban park green spaces in the PRD region. Utilizing the Relative Potency Factor method, the study calculates the Integrated Measure of Relative Potency Factors (IMI_{RPF}) for seven NEOs across various urban parks. It examines the possible relationship between these residues and regional economic development. Additionally, the study introduces the Direct Nemerow Index, a comprehensive evaluation method, to assess the health risks associated with exposure to NEOs in park green spaces. This research enhances our understanding of pesticide pollution in urban green space soils and offers valuable insights for the management and regulation of pesticides in parks.

2. Materials and methods

2.1. Chemicals and reagents

Analytical standards of seven NEOs—acetamiprid (99.75 %, purity), thiamethoxam (99.65 %), imidacloprid (98.55 %), clothianidin (99.12 %), thiacloprid (99.68 %), dinotefuran (99.25 %), and imidaclothiz (98.26 %)—were acquired from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Isotope-labeled standards imidacloprid-D4 (98.0 %) and clothianidin-D3 (99.9 %) were obtained from C/D/N Isotopes Inc. (Quebec, Canada). Acetonitrile and dichloromethane, employed in the extraction, were of analytical grade and sourced from Macklin Biochemical Co., Ltd. (China). Acetonitrile, used in the mobile phase, was of HPLC reagent grade, procured from Merck KGaA (Darmstadt, Germany). Primary secondary amine (PSA), a purifying agent, was sourced from Agela Technologies (China). Fig. S1 illustrates the molecular structures of the analyzed substances.

2.2. Sample collection

The PRD region is characterized by a subtropical monsoon climate.

The region's warm and humid conditions foster an ideal environment for pest growth and reproduction. Consequently, frequent pesticide use is necessary to manage this abundance of pests. Urban parks, essential components of urban greenery, play a crucial role in urban planning and landscape design. They enhance the living environment for residents and maintain the ecological balance within cities [17,18]. Urban parks, due to transport and proximity constraints, serve as popular spots for daily rest, weekend relaxation, and family leisure activities among city residents. In March 2023, we collected 427 soil samples from 85 parks within the PRD, with park types mainly including community parks and children's parks. Sampling locations were mapped via ArcGIS 10.2 (Environmental Systems Research Institute, USA) to Fig. 1. Depending on layout characteristics, 3 to 7 soil samples were gathered from each park. Sampling sites are primarily situated in regions of intense human activity, including both sides of major roads, special activity displays, grassy activity spaces, and camping zones. A single composite sample, around 500 g, was collected by mixing five soil samples (0–15 cm depth) from a 15 m × 15 m area. The samples were then transported to the lab and stored at −20 °C until analysis.

2.3. Sample extraction and instrumental analysis

Impurities such as leaves and stones were removed from the collected soil samples, which were then freeze-dried and sieved through a 0.212 mm mesh. The QuEChERS solid phase extraction method was selected for the pre-treatment of parkland soils. The pre-treatment was mainly referred to a fast and sensitive method for measuring multiple NEOs residues in soil [19]. Briefly, 5 g of pretreated soil was transferred into a polypropylene (PP) 50 mL centrifuge tube, followed by the addition of 7.5 mL of extractant (acetonitrile: dichloromethane at a 2:1 ratio). The tube was vortexed for 3 min, ultrasonically extracted for 15 min, and then centrifuged at 5000 r/min for 5 min, after which the supernatant was collected. This procedure was repeated twice, and the resulting supernatants were combined. The combined supernatants were transferred to 15 mL centrifuge tubes containing 200 mg of PSA, vortexed for 3 min, and then centrifuged at 5000 r/min for 5 min. Afterward, the supernatants were evaporated to dryness at 25 °C with nitrogen gas. The residues were vortexed for 3 min, then centrifuged at 5000 r/min for 5 min following an additional 10 min of vortexing. The residue was reconstituted in 200 µL of HPLC-grade acetonitrile by vortexing and filtered through a 0.2 µm nylon microcentrifuge filter (Anpel, China) into a 0.25 mL vial, ready for instrumental analysis.

2.4. LC-MS/MS instrumentation and operating condition

The instrument used was an HPLC-MS/MS (AB SCIEX, USA) equipped with a C18 column (100 mm × 4.6 mm, Agilent, USA). The injection volume was 1 µL. The mobile phase comprised two solutions: (A) water with 5 mM ammonium formate, and (B) acetonitrile. Chromatographic separation was performed at a flow rate of 0.4 mL/min using a gradient mobile phase as follows: 40 % B from 0 to 1 min, increasing linearly to 65 % B from 1 to 3 min, to 70 % B from 3 to 5 min, and to 97 % B from 5 to 6 min, before returning to 37 % B from 6 to 6.5 min and holding for 2 min. The MS system quantification was performed in multiple reaction monitoring (MRM) mode. Instrument acquisition parameters and related data are presented in Table S1.

2.5. Quality assurance and quality control (QA/QC)

Standard curves were plotted with gradient concentrations of 0.1, 0.5, 1, 2, 5, 10, 20, 50, 100, and 200 µg/L using imidacloprid-D4 (IMI-

D4) as the internal standard, with a consistent concentration of 50 ng/mL to monitor the instrument's operating status. Clothianidin-D3 (CLO-D3), an internal standard, was used to compensate for losses incurred during the pretreatment process. The efficiency of the pretreatment method was evaluated using three concentration levels (10, 50, 100 ng/mL). Recoveries ranged from 85–116.25 % in blank spiked samples and 73.97–103.73 % in matrix-spiked samples (soil without the target substance). CLO-D3 recoveries in matrix samples varied between 79.11 % and 81.98 %. All recoveries suggest that the procedure and condition of the instrument were acceptable. The limit of quantification (LOQ) was calculated as the mass corresponding to the lowest standard on the curve with a signal-to-noise ratio of 10, relative to the sample mass. The instrument LOQs ranged from 0.17 to 2.27 µg/L for each substance. In every batch of samples, a blank program sample, a blank spiked sample, and two parallel samples were also measured simultaneously.

2.6. Statistical analyses

Statistical analyses were performed using SPSS 20.0 (IBM Corp., Armonk, NY, USA). Non-parametric test indicators were assessed using the Mann-Whitney U test, with a significance level set at 0.05. The spearman correlation coefficient > 0.8 was considered a strong correlation.

2.7. Relative potency factor method

For comparing regional differences in ΣNEOs concentrations, detected NEOs were standardized against a common reference. Imidacloprid, the most frequently detected and extensively studied NEOs, was selected as the index compound[20]. The RPF calculation, presented in Eq. (1), is based on the acceptable reference dose (RfD) provided by the U.S. Environmental Protection Agency (EPA)[21] as shown in Table S2, leading to the final calculation in Eq. (2). Since imidaclothiz lacks an associated RfD, its value was aligned with that of imidacloprid. For samples with residue levels below the LOD, half of the LOD value was utilized in the RPF calculation.

$$RPF_i = RfD_i / RfD_{imidacloprid} \quad (1)$$

$$\begin{aligned} IMI_{RPF} &= \sum_i^n (\text{neonicotinoid} \times RPF_i) \\ &= \text{imidacloprid} + \text{dinotefuran} \times 2.85 + \text{acetamiprid} \times 0.803 \\ &\quad + \text{thiacloprid} \times 14.25 + \text{clothianidin} \times 5.816 \\ &\quad + \text{thiamethoxam} \times 9.5 + \text{imidaclothiz} \end{aligned} \quad (2)$$

2.8. Time-Cumulative toxicity of NEOs and application of direct Nemerow pollution index

Traditional chemical toxicity studies focus primarily on the concentration-effect model, which determines a chemical's toxicity or safety by observing its LC50 (EC50) or no-observed-adverse effect level (NOAEL) in various model organisms throughout the exposure cycle. According to Haber's rule ($K = C \times T$), emphasizing concentration effects without consideration of exposure duration introduces bias. In "Dosis und Wirkung" (Dose and Response), Druckrey and Küpfmüller discuss two determinants of poison toxicity: the exposure concentration/dose in the key receptor and the ratio of the dissociation time constant (TD) to the association time constant (TA), reflecting the dynamics of poison-receptor interactions. When TA is significantly less than TD, this indicates a cumulative effect of the toxicant concentration/dose in the key receptor after repeated exposures, signifying time-dependent toxicity [22].

NEOs, systemic insecticides that bind strongly and irreversibly with nAChRs, are believed to exhibit potential time-dependent toxicity. This has been explored using the time to event (TTE) method[23] and confirmed in empirical studies. In researching the time-dependent toxicities of prevalent pesticides in Israeli honey, Bommuraj uncovered evidence for the time-reinforced toxicity of acetamiprid, as demonstrated by a significant negative regression between log (LC50) and log (time), indicating increased toxicity over time[24,25]. However, the RfD values for NEOs' toxicity are derived from the concentration-effect model. Soil exposure risk assessments also rely on this model, comparing exposure doses to RfD and treating residual concentrations as indicative of acute toxicity, thus overlooking NEOs' time-dependent toxicity. The presence of NEO residues in soil, influenced by its properties, migration, transformation, and the variable nature of contamination sources, necessitates a sophisticated assessment approach that transcends reliance on average data values for accurate evaluation of their acute and chronic toxicity in a specific region.

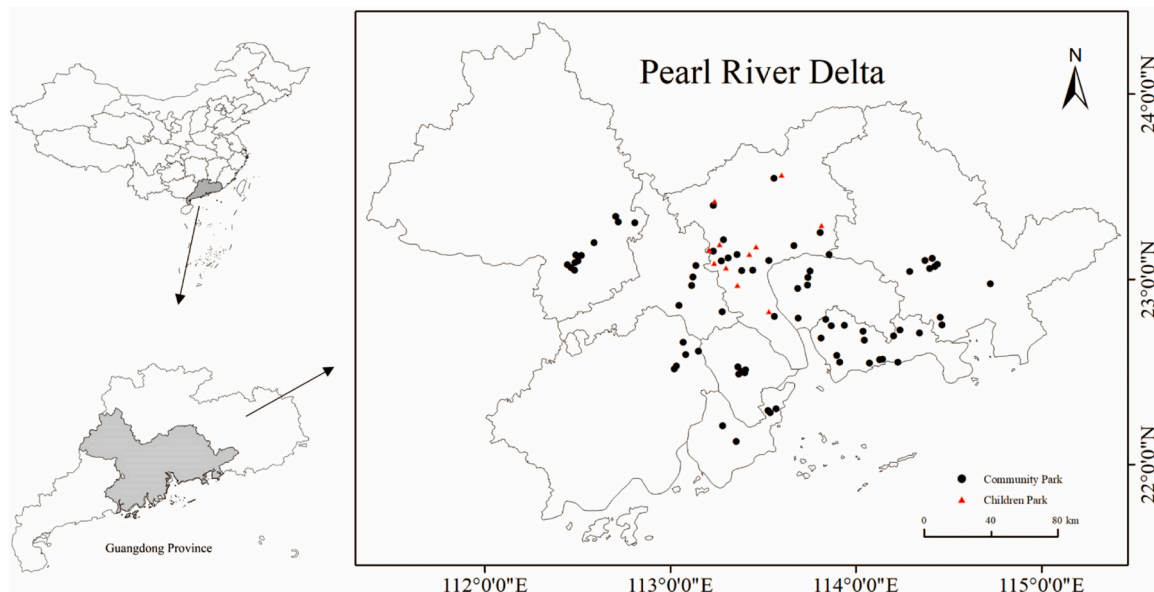


Fig. 1. Map of Sampling Sites of Urban Parks in the PRD.

The Nemerow index method evaluates environmental quality by considering both average and extreme values, presented in Eq. (3) [26, 27]. This method is a weighted multifactorial measure initially created to assess environmental multi-pollutant levels by considering both average exceedances and the most extreme pollution events. Owing to the lack of environmental background values for NEOs and their distinct acute and chronic toxicities, we formulated a novel, direct NPI definition to analyze and assess NEOs residue distributions in parks. For NEOs residues, we propose defining the maximum value as indicative of short-term acute exposure and the average value as reflective of long-term chronic exposure. This facilitates the direct application of the Nemerow index to comprehensively assess environmental exposure levels. By considering the residue distribution and incorporating both mean and maximum values, we can provide a more scientifically sound evaluation of the Toxicity Residue Value in parks resulting from NEOs toxicity. Furthermore, this method enables an effective comparison of NEOs distribution differences. This methodology could also be used to compare NEOs residue levels across various regions within the same medium, with further research expected on its application.

$$NPI = \sqrt{\frac{P_{ave}^2 + P_{max}^2}{2}} \quad (3)$$

Where NPI is Nemerow pollution index, P_{ave} is the average contamination index, P_{max} is the Maximum contamination index.

2.9. Cumulative exposure probability risk assessment model

The non-carcinogenic human health risk from NEOs exposure was assessed using the US EPA's health risk evaluation model, which considers three exposure pathways to contaminated soil: ingestion, dermal contact, and inhalation, presented in Eq. (4-6), with the parameters shown in Table S3. Probability distributions for NEOs residues were generated using Monte Carlo simulations (Crystal Ball software, Oracle Inc., CA, USA), providing predicted exposure dose distributions for each pathway.

(1) Soil Ingestion

$$ADD_{soil\ ingestion} = C_i \times IRS \times EF \times ED / BW / AT \times 10^{-6} \quad (4)$$

(2) Dermal Contact

$$ADD_{dermal} = C_i \times SA \times AF \times EF \times ED \times ABS / BW / AT \times 10^{-6} \quad (5)$$

(3) Breath Inhalation

$$ADD_{inhalation} = C_i \times ED \times EF \times PM_{10} \times DAIR \times PLAF \times FSPO / BW / AT \times 10^{-6} \quad (6)$$

where C_i is the concentration of NEOs in the soil ($\text{mg} \cdot \text{kg}^{-1}$), IRS is the soil ingestion rate ($\text{mg} \cdot \text{day}^{-1}$), ED is the exposure duration (years), EF is the exposure frequency (days/year), BW is the body weight (kg), AT is the mean exposure time (days), SA is the skin surface area (cm^2), AF is the skin adherence factor ($\text{mg} \cdot \text{cm}^{-2}$), ABS is the dermal absorption factor [28], PM_{10} is the inhalable particle matter ($\text{mg} \cdot \text{m}^{-3}$), $DAIR$ is the daily aerial respiration ($\text{m}^3 \cdot \text{d}^{-1}$), $PLAF$ is the proportion of inhaled soil particles retained in the body, and $FSPO$ is the proportion of particles from the soil in the air.

To calculate the risk of population exposure, the residue probability distributions of NEOs residues in all parks calculated by the direct Nemerow's exponential method were fitted using Monte-Carlo simulation, and the NEOs were normalized by using the IMI_{RPF} method to

calculate the normalized daily dose of NEOs exposure for three exposure routes of different populations, and the distribution of cumulative probability risks for different routes of exposure was obtained for both adults and children. The normalized daily dose of NEO exposure depends on both residual concentration and RfD .

3. Results and discussion

3.1. The residues of Σ_7 NEOs in the urban park green space

The residual levels of Σ_7 NEOs in the soil of urban park green spaces in the PRD region are presented in Table 1. Among the 472 soil samples, the detected concentrations of NEOs were ranked from highest to lowest as follows: imidacloprid > clothianidin > acetamiprid > thiamethoxam > dinotefuran > thiacloprid > imidaclothiz. The detection rates ranged from 0 to 89.23 %, with the predominant NEOs being imidacloprid and clothianidin, which had detection rates of 89.23 % and 83.37 %, respectively. At concentration levels, imidacloprid and clothianidin were the predominant residues, with ranges of (N.D.-87.91, 3.58 $\mu\text{g}/\text{kg}$) and (N.D.-60.67, 1.57 $\mu\text{g}/\text{kg}$) respectively, forming the majority of the mean concentration. The secondary residues were acetamiprid and thiamethoxam ranged from (N.D.-113.19, 0.69 $\mu\text{g}/\text{kg}$) to (N.D.-48.16, 0.39 $\mu\text{g}/\text{kg}$).

The climate of the PRD, characterized by mild temperatures, high humidity, and short winters, promotes year-round pest activity, leading to significant pest damage [29]. The high detection rates indicate the widespread use of NEOs in PRD parks for pest control and plant protection. The predominant NEOs used in this context are imidacloprid and clothianidin, which aligns with the findings of previous studies [30-32]. Consequently, NEOs have been extensively utilized for pest management in urban park green spaces within the PRD region.

When compared to similar research in Guangdong Province, Yu et al. [9] reported average NEOs residues in Guangzhou City's agricultural soil (collected from April to June) to be 24 $\mu\text{g}/\text{kg}$ (maximum of 370 $\mu\text{g}/\text{kg}$), an order of magnitude higher than our findings. The average concentrations of clothianidin and acetamiprid were 6.2 and 2.0 $\mu\text{g}/\text{kg}$ (maxima of 70 and 20 $\mu\text{g}/\text{kg}$), respectively, higher than those in our study. Gu et al.'s large-scale study [33] of farmland soil residues in Guangdong Province revealed mean concentrations of imidacloprid, clothianidin, acetamiprid, and thiamethoxam ranging from 0.52 to 6.63 $\mu\text{g}/\text{kg}$ (maximum values between 1.49 and 21.00 $\mu\text{g}/\text{kg}$), lower than Yu's results but still exceeding our own. Comparative studies of NEOs across various regions in Guangdong Province have indicated that the residual levels in agricultural soils surpass those in typical non-agricultural lands-park green spaces. However, with an outlier value of 113.19 $\mu\text{g}/\text{kg}$ for acetamiprid in our study, the residues of parkland soils occasionally exceeded those in farm soils. The presence of NEOs residues is noteworthy and may be associated with horticultural activities specific to parks, which differ from those in agricultural areas [30].

Table 1

The mean, maximum, and median values of detected NEOs concentrations (ng/g) and detection frequency (DF) in samples.

| NEOs (n = 427) | Mean | Max | Median | DF% |
|----------------|------|--------|--------|-------|
| Imidacloprid | 3.58 | 87.91 | 0.37 | 89.23 |
| Acetamiprid | 0.69 | 113.19 | N.D. | 52.22 |
| Dinotefuran | 0.01 | 1.78 | N.D. | 4.92 |
| Imidaclothiz | N.D. | N.D. | N.D. | 0.00 |
| Clothianidin | 1.57 | 60.67 | 0.17 | 83.37 |
| Thiacloprid | 0.00 | 0.04 | N.D. | 1.41 |
| Thiamethoxam | 0.39 | 48.16 | N.D. | 43.56 |
| Σ NEOs | 6.25 | 137.31 | 0.83 | 93.91 |

3.2. Comparison of NEOs residues in urban green space of different park types

To investigate the distribution characteristics of NEOs in different types of parks, we selected soil samples from various park green spaces in Guangzhou, a typical city in the PRD region, for classification analysis. Table 2 shows that imidacloprid and clothianidin had high and similar detection frequency in both park types (93.84 %–98.18 %). The distinction lies in the fact that acetamiprid and thiamethoxam were significantly more detected in children’s parks compared to community parks. For the more intuitive comparison, we plotted a cumulative histogram (Fig. 2) that represents the average NEOs concentrations in soils from each park by computing the mean concentrations from five soil samples per park, thereby reducing the individual samples from 55 to 11 representative cumulative bars. This approach ensures that variations within individual parks do not overshadow the overall comparison between park types. A comparison of mean, maximum, and median values revealed that NEOs residue levels in children’s parks were higher relative to community parks. The histogram reveals that overall NEOs concentrations are generally higher in children’s parks than in community parks. The Mann-Whitney U test confirmed a significant difference in NEOs concentrations between the types of parks ($p < 0.01$). Overall, the residues of NEOs were generally found to be higher in children’s parks than in community parks, both in overall levels and for individual compounds.

As a city with a high level of economic development in the PRD region, Guangzhou has been at the forefront of developing China’s largest children’s park system since 2015. Given the unique vulnerability of children, the safety of their play in these parks has garnered heightened attention, potentially leading to more stringent park management practices, including pest control measures. This increased focus on safety and management might explain the significantly higher levels of NEOs residues observed in children’s parks compared to community parks. In response, we have provided relevant descriptions in the discussion section of the original text.

3.3. Integrated NEOs concentrations (IMI_{RPF}) in urban green space

Considering the similar toxicological endpoints and mechanisms of NEOs, we utilized the IMI_{RPF} method to consolidate their toxicity into a composite index. The IMI_{RPF} is visually depicted using box-and-whisker plots (Fig. 3). The box plot analysis shows that the Zhuhai and Guangzhou regions display a wider spread of NEOs residues. Considering the outliers, a small proportion of samples from Shenzhen, Huizhou, Guangzhou, and Dongguan show high NEOs levels. Regarding mean values, economically advanced areas like Shenzhen (29.16), Guangzhou (19.05), and Zhuhai (19.30) yield higher figures. The residues of NEOs in park green space are largely due to human-initiated pest control measures. Differences in the frequency of NEOs applications, the variety of compounds used, and the rate at which these compounds degrade and migrate through park soils may lead to significant variations in their distribution. This is evident from the numerous outliers observed in the chart, which highlight the diverse patterns of NEO

Table 2

The mean, maximum, and median values of detected NEOs concentrations ($\mu\text{g}\cdot\text{kg}^{-1}$) and detection frequency (DF) in Children Park and Community Park.

| Park type | NEOs | Mean | Max | Median | DF% |
|-------------------------|--------------|------|-------|--------|-------|
| Children Park (n = 55) | Imidacloprid | 7.70 | 60.32 | 3.20 | 96.36 |
| | Acetamiprid | 2.17 | 32.50 | 0.06 | 58.18 |
| | Clothianidin | 3.13 | 48.03 | 0.27 | 98.18 |
| | Thiamethoxam | 0.33 | 3.23 | 0.05 | 58.18 |
| | Imidacloprid | 2.22 | 17.76 | 0.50 | 96.92 |
| Community Park (n = 65) | Acetamiprid | 0.02 | 0.31 | N.D. | 23.08 |
| | Clothianidin | 0.88 | 12.91 | 0.25 | 93.84 |
| | Thiamethoxam | 0.17 | 3.25 | N.D. | 38.46 |

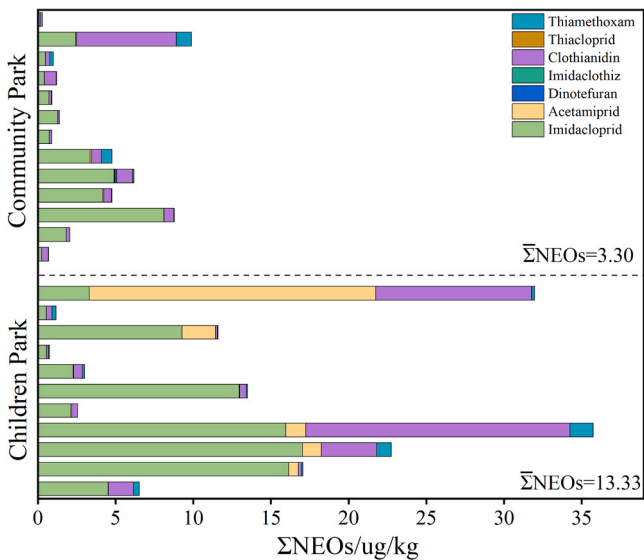


Fig. 2. Cumulative Histogram of NEOs Residues in Soil from Children Parks and Community Parks.

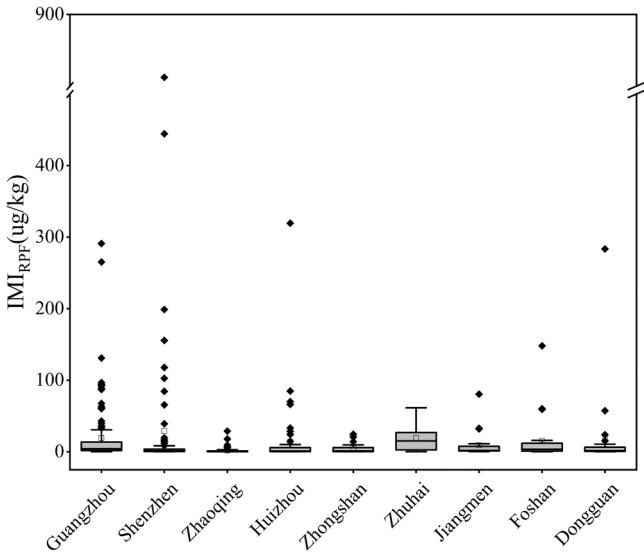


Fig. 3. Box plots of the IMI_{RPF} by region.

presence across different park settings.

To explore the potential factors influencing the distribution differences of NEOs residues in park green spaces across cities within regions, we have initially examined the potential link between economic factors and the levels of NEOs residues in urban green soil. Considering regional differences in area, population, and economic scale, we used per capita GDP as a metric to examine its correlation with the IMI_{RPF} . As depicted in Fig. 4, a fitted linear curve indicates a good correlation ($R^2 = 0.86$), suggesting that areas with greater economic development may exhibit higher NEOs residue levels. The deviations towards higher values in the figure may stem from the limited representativeness and number of parks, amplifying outlier effects and skewing results. Conversely, the lower deviation IMI_{RPF} values of Zhaoqing may be due to its slower urbanization and economic growth, with public service investment still evolving. Furthermore, rich forest resources in Zhaoqing provide ample green space for its residents, reducing the need for additional parklands, reflected in the lower IMI_{RPF} values.

The pace of economic development and urbanization in the PRD region differs among cities. As urbanization progresses, the urban

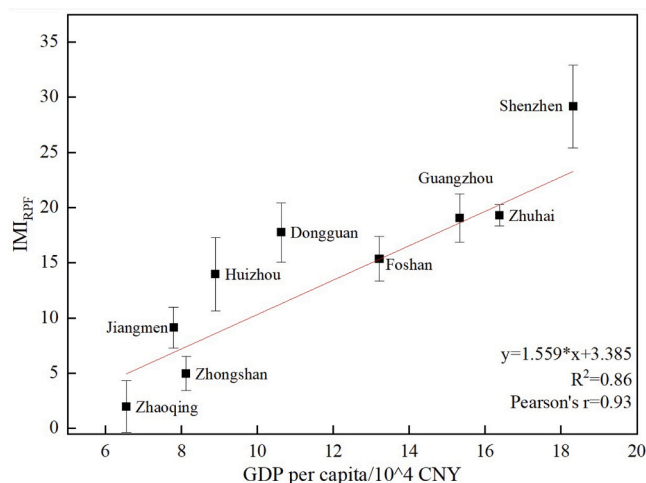


Fig. 4. Scatterplot of correlation between log-transformed IMI_{RPF} and regional GDP per capita (error bars chosen as coefficient of variation).

landscaping industry, a key component of city construction, has experienced rapid growth. The primary source of funding for the maintenance of green spaces in urban areas is the government's urban maintenance budget. A higher demand for urban parkland often leads to increased government investment in their construction and management. This heightened expenditure on park development and upkeep can result in significant variations in the use of NEOs and the distribution of their residues in parks. This phenomenon may account for the observed positive correlation between per capita GDP and the concentration of NEOs residues in these areas.

3.4. Health risk assessment of NEOs in urban green space

In this study, we identified four primary NEOs in the soil of urban park green spaces in the PRD region: imidacloprid, clothianidin, acetamiprid, and thiamethoxam. Focusing on these substances, we calculated the daily cumulative exposure doses for various populations across three exposure pathways: soil ingestion, dermal contact, and inhalation, as depicted in Fig. 5. The results indicate that the estimated combined NEOs exposure doses for both adults and children in parks, even at cumulative probabilities approaching 100 %, remain below $1 \mu\text{g/kg/day}$, which is notably lower than the EPA's RfD for imidacloprid at $57 \mu\text{g/kg/day}$. Notably, children's daily exposure doses are approximately an order of magnitude higher than those of adults. The risk ranking of the three exposure pathways is as follows: Soil Ingestion > Dermal Contact

> Inhalation. The contribution of individual NEOs to exposure risk is as follows: clothianidin (40.23 %), imidacloprid (30.88 %), thiamethoxam (24.44 %), and acetamiprid (4.45 %), reflecting the interplay between residue levels and toxic risk.

Based on these, we can conclude that the combined NEOs exposure risk faced by urban populations in the PRD region in park scenarios is still at an acceptable level in terms of long-term exposure. However, it is important to note that due to some abnormally high values obtained in residue detection, acute exposure from short-term NEOs application should receive more emphasis in park management activities. In our actual survey, we found that parks were often closed for 1–2 weeks to reduce the risk of pesticide exposure under seasonal pest control, which effectively reduced the acute toxicity of NEOs to the visiting population. Overall, the risk of combined NEOs exposure to urban populations in park settings in the PRD region is deemed acceptable.

4. Conclusion

In the PRD, the primary NEOs detected in park green spaces are imidacloprid and clothianidin, with detection rates of 89.23 % and 83.37 %, respectively. The concentrations of NEOs in the soil vary from 0.83 to $137.31 \mu\text{g/kg}$. Notably, in Guangzhou, children's park green spaces show significantly higher levels of NEOs residues compared to community parks. Through inter-city comparisons using the IMI_{RPF} , a positive correlation was established between cities IMI_{RPF} values and per capita GDP, ($R^2 = 0.86$). This finding suggests that the levels of NEOs in urban environments may be linked to economic development. In conclusion, the risk of urban populations in the Pearl River Delta region being exposed to NEOs in park environments is considered acceptable.

This study is beneficial for understanding the residual status of NEOs in urban green spaces, which is crucial for protecting human health and fostering sustainable ecological development. The results emphasize the importance of monitoring and managing pesticide use in urban areas to mitigate the potential risks associated with NEO exposure.

Environmental implication

Neonicotinoids (NEOs) are a specific type of chemical insecticides. The extensive use of NEOs, along with their irrational prophylactic application patterns, has led to their prevalence as pollutants in various environments. Since NEOs were linked to bee deaths in 2013, concerns have grown over their risks to non-target organisms and humans.

This study systematically investigated the prevalence of seven NEOs insecticides in urban parklands, and assessed the health risks to residents. We explored connections between pollution and economic growth and employed the direct Nemerow index for a comprehensive

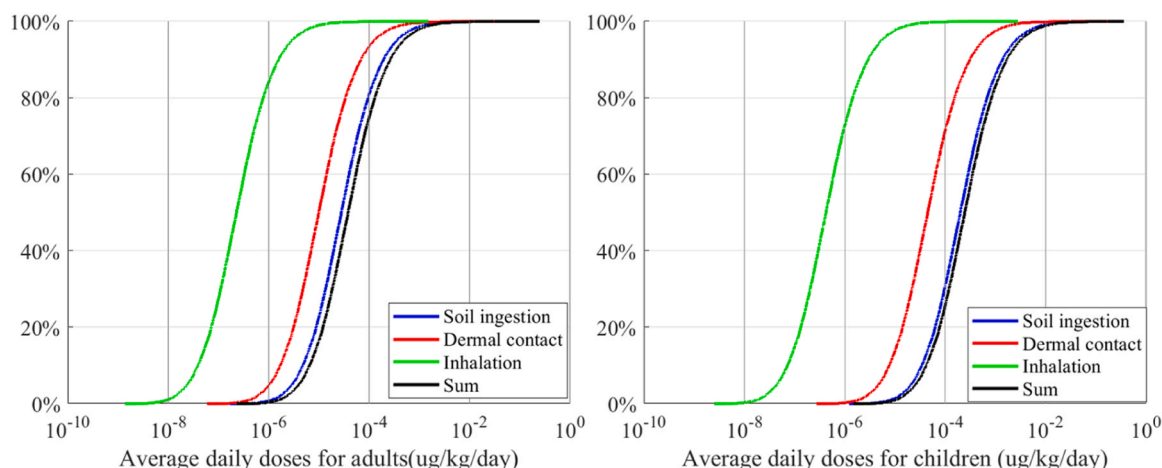


Fig. 5. The cumulative distributions of the estimated average daily doses (ADDs) of IMI_{RPF} from soil ingestion, dermal contact, inhalation and sum of them.

residue assessment.

CRediT authorship contribution statement

Guocheng Hu: Supervision. **Pengchong Wen:** Methodology. **Ping Ding:** Project administration. **Limei Cai:** Resources. **Yunjiang Yu:** Resources. **Haojie Luo:** Writing – original draft, Methodology. **Jun Pan:** Investigation. **Yanan Sun:** Validation, Investigation.

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

The data that has been used is confidential.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jhazmat.2024.135330](https://doi.org/10.1016/j.jhazmat.2024.135330).

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