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Residues of neonicotinoid insecticides in surface sediments in lakes and rivers across Jiangsu Province: Impact of regional characteristics and land use types☆

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ABSTRACT

Neonicotinoid insecticides (NNIs) had been detected in soil and surface water frequently because of extensive use worldwide, however, data regarding regional characteristics and potential influential factors of sediment were scarce. In the present study, eight NNIs were analyzed in 86 surface sediment samples from different regions (central cities, rural areas and suburbs) and land use types (construction land and crop land) in Jiangsu Province. NNIs were widespread in the sediments, with a mean value of 1.73 ± 0.89 ng g^{-1} dry weight (dw) (ranged from 0.41 to 3.87 ng g^{-1} dw). Imidaclothiz (IMIZ), dinotefuran (DIN) and nitenpyram (NIT) were the dominant compounds in the surface sediment, accounted for half of combined total. The results of regional distribution analysis show that NNIs were at higher concentrations in rural areas and crop land, while the residues of NNIs in lakes were more severe compare with rivers in Jiangsu Province. Region characteristics and land use types have an influence on residues of NNIs in surface sediment. Principal component analysis showed that residues of NNIs in surface sediment in Jiangsu Province mainly originated from protect grain crops (maize), fruit (apples, pears) and vegetables in agricultural systems. The residues of NNIs were found to be mostly concentrated in the northwest and northeast in Jiangsu Province, where were the area of intensive agriculture. To investigate the residues of NNIs, while identify the contributing factors, could provide a scientific basis for basic of region environment management and pollution control.

1. Introduction

Neonicotinoid insecticides (NNIs) are one of the most widely used insecticides class worldwide, which have been currently registered and approved for use in more than 120 countries (Main et al., 2015), comprising about 25% of the world market of insecticides (Simon-Delso et al., 2015; Zhou et al., 2021). Due to its high selectivity, good bioavailability and low toxicity to vertebrates, NNIs are applied in all kinds of crops, forestry, plant nurseries, gardens, urban parks, veterinary products, etc. (Bonmatin et al., 2015; Bonmatin et al., 2019; Kagabu, 2011; Matsuda et al., 2005; Wang et al., 2019). In recent years, NNIs were frequently detected in environment (e.g., soil, dust, surface water, sediment) and food contamination (e.g., vegetable, fruit, tea) (Forero et al., 2017; Huang et al., 2020; Jiao et al., 2016; Schaafsma et al., 2015; Zhou et al., 2021). Besides, several NNIs have been proved to possess sublethal to lethal effects on bees, butterflies, crustaceans, fish, bobwhites, mallards, rat (Wang et al., 2018; Zhang et al., 2020), and been shown to perturb the endocrine system of lizards (Wang et al., 2020), which revealed NNIs posed potential risks to human health and ecosystems. In Europe, clothianidin (CLO), imidacloprid (IMI) and thiamethoxam (THM) had been gradually limited their applications in outdoor in 2013 (EC, 2013a; 2013b), thiacloprid (THA) was completely disabled in 2020 (EC, 2019), and NNIs was listed into an observation checklist of surface waters in 2015 (EC, 2015). Contrastingly, the use of

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pesticide (insecticides mainly) in China increased drastically in the last decade, which emerged as the foremost producer, consumer and exporter of pesticides (FAO, 2020). Therefore, the potential risks caused by insecticides should be attracted more attention in China.

With high water solubility, low soil affinity and low uptake rates of crops (approximately 5%) (Lewis et al., 2016; Zhang et al., 2018), NNIs are easily migrated into aquatic environment through surface runoff (Hladik et al., 2018; Morrissey et al., 2015). Numerous evidence is now available on the ubiquity of NNIs in surface water, with concentrations even exceeding the aquatic benchmarks for acute exposure in some cases (Anderson et al., 2015; Hladik and Kolpin, 2016; Yamamuro et al., 2019). However, studies about NNIs in sediment were limited thus far, though they might be adsorbed in sediment and entailed a threat not only for benthic species but also for feeding on benthic and demersal communities (Bonmatin et al., 2019; Zhang et al., 2019).

In order to better understand the occurrence and source of NNIs in sediment, it is essential to elucidate the main influential factors for their spatial distribution. It had been suggested that the most significant factor influencing the composition of pesticides enriched in sediment was the application patterns (Li et al., 2014; Wei et al., 2017). For example, acetamiprid (ACE) and CLO were the dominant NNIs in sediment in Guangzhou (Zhang et al., 2020). IMI was the main pollution substance in sediment from rice planting area in Fujian, whereas IMI, ACE and THM were the most abundant NNIs in sediment from vegetable crop area in Dongguan (Huang et al., 2020). In Belize, the main component of NNIs in sediment was IMI (Bonmatin et al., 2019). Therefore, it might be a significant route to understand the source of NNIs via surveys with regional characteristics and land use types around river channels. To date, there is no report on NNI residues in sediment locating in suburbs, and information on NNI residues in sediment located in region of different land use types are limited.

Jiangsu Province is one of the most economically developed coastal provinces and the second largest province in gross annual value of industrial and agricultural output in China, with a dense river network and numerous lakes. According to the statistics, the yield of chemical pesticide reached 538 thousand tons, which were produced by 28 major pesticide enterprises in 2017 in Jiangsu Province. The total production of neonicotinoids was 93 thousand tons, which was a 14.6% increase from the preceding year (Chemical report network, 2018). A more intensive use of pesticide (neonicotinoids mainly) may pose even greater risks to the aqueous environment in Jiangsu Province.

Consequently, the present study took Jiangsu Province as the research area, and aimed to reveal the spatial distribution characteristics and understand the influence of different regions and land use types on NNIs. Firstly, the components and contents of NNIs were analyzed in surface sediment samples, which were collected from 12 lakes and 10 rivers in Jiangsu Province. Secondly, the relationship between distribution of NNIs in surface sediment, regional characteristics and land use types were explored. Lastly, Principal component analysis (PCA) was performed to identify source apportionment of NNIs based on environmental metrics.

2. Materials and methods

2.1. Chemicals

Eight targeted individual neonicotinoid compounds, including IMI, ACE, nitenpyram (NIT), imidaclothiz (IMIZ), THA, THM, CLO, dinotefuran (DIN) (purity: 98.0%–99.9%), and two isotope-labeled standards (IMI-d_4 and CLO-d_3 (purity: 98.0%–99.9%)) were purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Acetonitrile (HPLC-MS grade) was purchased from CNW Technologies GmbH (Dusseldorf, Germany). Anhydrous $MgSO_4$ and NaCl were obtained from ANPEL Laboratory Technologies (Shanghai) Inc. (Shanghai, China).

2.2. Sampling collection

This study covered almost different rivers and lakes of the Jiangsu Province, including 12 lakes (e.g., Tai Lake, Yangcheng Lake, Dianshan Lake, Ge Lake, Changdang Lake, Shijiu Lake, Shaobo Lake, Gaoyou Lake, Baima Lake, Hongze Lake, Chengzi Lake, Luoma Lake) and 10 rivers (e.g., Beijing-Hangzhou Canal, Yangtze River, Tonglv Canal, Tongyang Canal, Tongyu Canal, Huai River, Sheyang River, Old Yellow River, Shu River, Yi River).

A total of 86 surface sediment samples were collected from Jiangsu Province between December 2019 and January 2020 (Fig. 1), according to the different watersheds, regions (e.g., rural areas, central cities and suburbs) and land-use types (e.g., crop land and construction land) (Table S1). The surface sediment samples were sieved on site to remove leaves and stones after collection, and then collected into polypropylene tubes. All samples were stored at −20 °C after collection until being assayed.

2.3. Sample extraction and instrumental analysis

The surface sediment samples were lyophilized and milled to pass through a 100 mesh (150 μm pore size) screen, which were then extracted by liquid–liquid extraction. After adding 5.0 g of freeze-dried sediment sample, the surrogate standards (CLO-d_3) and 20 mL mixtures of acetonitrile and dichloromethane (v/v, 2:1) were added into a 50 mL polypropylene centrifuge tube. The sediment slurry was vortexed for 10 min at 2200 rpm, sonicated for 15 min at 30 °C, centrifuged for 8 min at 4000 rpm, and then decanted the supernatant. The extraction steps were repeated once, and the supernatants were combined. The supernatant was evaporated to dryness and re-dissolved in 1.0 mL acetone and transferred with 10 mL Milli-Q water in a 50 mL polypropylene centrifuge tube. A 2.0 g NaCl and 5 mL dichloromethane were added in the tube, followed by vortexing for 5 min (2200 rpm) and centrifuging for 5 min (4000 rpm). The supernatant was transferred to a new 15 mL polypropylene centrifuge tube, dried by nitrogen blow, and resuspended in 1 mL acetonitrile. The extracts were filtered through a 0.2 μm nylon filter to the LC vial and brought to an Agilent 1260 LC/AB Sciex 4000 Qtrap MS (LC-MS/MS, Agilent Technologies, Santa Clara, CA, USA) analysis. 50 μL of a 1 μg/mL solution of internal standard (IMI-d_4) was added to the LC vial prior to instrumental analysis.

LC-MS/MS was operated in the negative mode with electrospray ionization. A CORTECSTM C18 column (4.6 × 100 mm; 2.7 μm, Waters, USA) was used for the separation of NNIs at 40 °C. The mobile phase consisting of acetonitrile (A) and 1% formic acid aqueous solution (B) was pumped at a flow rate of 0.4 mL/min. Gradient elution scheme for high-performance liquid chromatography and parameters of tandem mass spectrometry are shown in Table S2.

2.4. Quality assurance and quality control

The quality control system, including procedural blank, laboratory blank, matrix blank and matrix spiked samples were used to validate the precision and accuracy of the experimental method before chemical analyses. The instrument was calibrated using external standard before analysis to ensure the stability of performance (within 20% variation of the individual compounds). The blanks samples for all target neonicotinoids were below the method detection limit (MDL). The recovery for the target insecticides from 78.3% to 123% in the matrix spiked samples. The recoveries of the internal standards were 81.5%–122%. Quantification was based on internal calibration standard 8 points curve ($R^2 > 0.999$, range between 1 and 200 μg/L) (Table S3).

2.5. Statistical analysis

Principal Component Analysis (PCA) with varimax rotation was carried out to determine the relationship between the NNIs and to

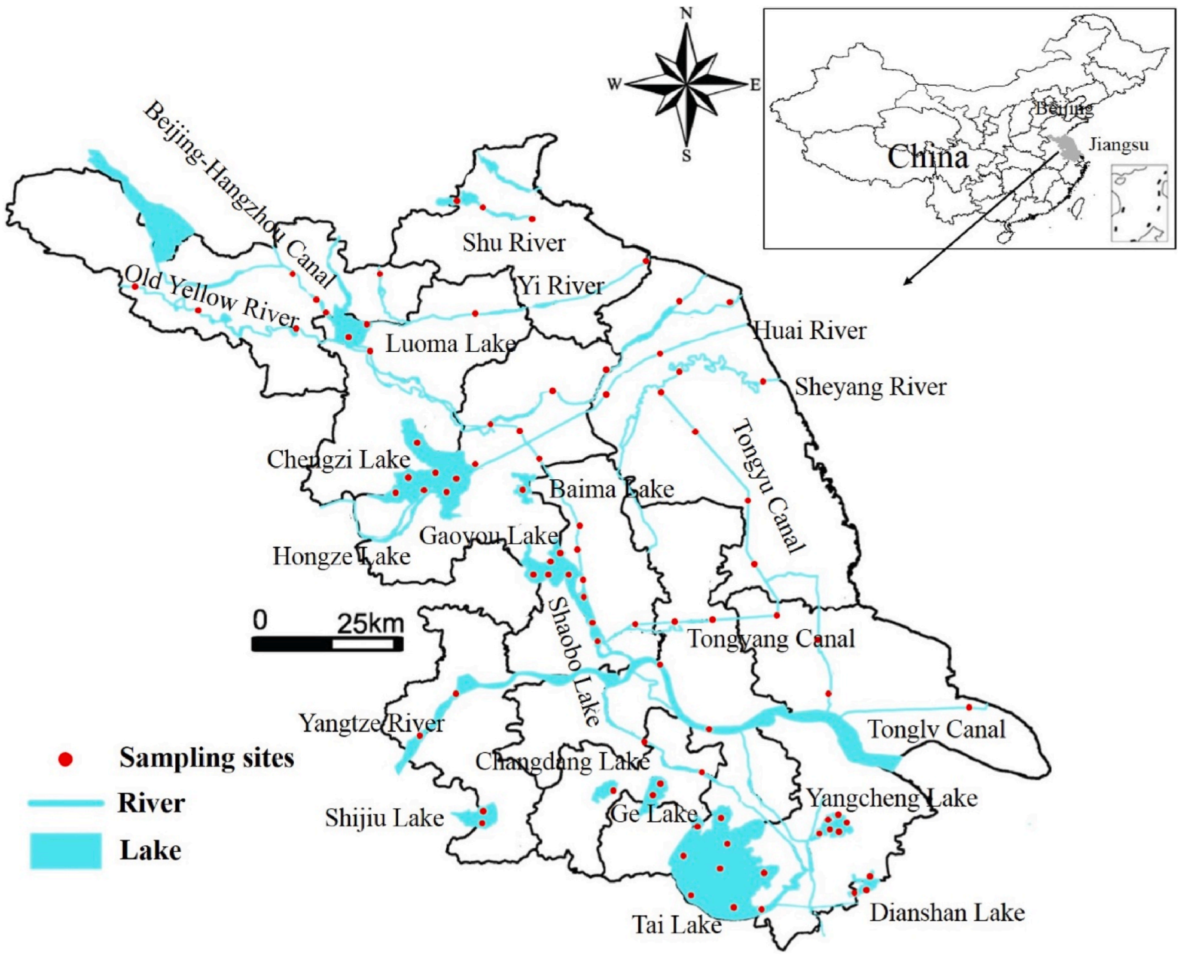

Fig. 1. Map of the surface sediment sampling sites in Jiangsu Province.

identify the possible origins of the NNIs. The Mann-Whitney *U* test was used for testing differences. SPSS 18.0, Origin 9.1 and ArcGIS 10.4.1 were used for data handling, statistical analyses and graphical outputs.

3. Results and discussion

3.1. Occurrence and distribution of NNIs in surface sediment

The concentrations and detection frequency (DF) of eight NNIs in surface sediment in Jiangsu Province are summarized in Table 1. The DFs were 100% for six NNIs, including IMI, NIT, IMIZ, THA, CLO and DIN, whilst those for ACE and THM were 97% and 95%, respectively. The ΣNNIs levels ranged from 0.41 to 3.87 ng g^{-1} dry weight (dw) with a mean value of 1.73 ± 0.89 ng g^{-1} dw in the 86 sediments. Among the eight NNIs, IMIZ was the highest arithmetic mean value (0.35 ng g^{-1} dw), followed by DIN (0.30 ng g^{-1} dw), NIT (0.25 ng g^{-1} dw), IMI (0.22 ng g^{-1} dw), THM (0.19 ng g^{-1} dw), THA (0.18 ng g^{-1} dw), CLO (0.16 ng g^{-1} dw) and ACE (0.09 ng g^{-1} dw). This may stem from the higher activity (20 fold or higher than ACE and IMI), broad spectrum, low cost and not affected by high or low temperature in IMIZ.

Regarding NNIs composition, IMIZ (23.08%), DIN (15.69%) and NIT (14.14%) dominated the total NNIs pollution, more than half of combined total. IMI, THM, THA, CLO and ACE contributed 11.81%, 11.12%, 10.00%, 8.73%, and 5.43%, respectively. The main component of NNIs here displayed varied across those collected from Belize and Guangzhou (Bonmatin et al., 2019; Zhang et al., 2020), in which THM and ACE were the main component, and the arithmetic mean value of the NNIs (including IMI, ACE, THA, THM and CLO) was 0.48 and 0.25 ng g^{-1} dw, respectively, far below the values in this study. It might be interpreted by the use of insecticide. Jiangsu Province is the developed area of intensive agriculture in China, where needs more use of neonicotinoids. It had been reported that the frequency of use of DIN and NIT had dramatically decreased in 5–10 years ago, and the production of IMIZ was relatively small (only 200 tons per year) before 2010 (Chen et al., 2019a). Accordingly, the contamination of IMIZ, DIN and NIT should be low. However, IMIZ, DIN and NIT in the present study were found to be higher than those more widely and largely used compounds (e.g., IMI and ACE), indicating possible shifts on the type of neonicotinoids being applied. This is consistent with the results found in a previous study

Table 1
Detection frequency (DF) and concentrations of NNIs in surface sediment samples in Jiangsu Province.

Compounds	DF(%)	Concentration (ng·g^{-1} dry weight)			
		Mean	Standard deviation	Max	Min
IMI	100	0.22	0.26	1.38	0.0495
ACE	97	0.09	0.13	0.82	<RL[a]
NIT	100	0.25	0.23	1.20	0.0296
IMIZ	100	0.35	0.34	2.00	0.0488
THA	100	0.18	0.32	1.34	0.0004
THM	95	0.19	0.33	2.40	<RL
CLO	100	0.16	0.14	1.00	0.0480
DIN	100	0.30	0.31	1.43	0.0045
∑NNIs[b]	100	1.73	0.89	3.87	0.4100

[a] Less than the reporting limit (RL).
[b] The sum of the concentrations of IMI, ACE, NIT, IMIZ, THA, THM, CLO and DIN in surface sediments.

(Chen et al., 2019a). The contamination profiles of NNIs in sediment varied by region might be related to their usage patterns.

The concentrations of eight NNIs in surface sediments in lakes and rivers across Jiangsu Province were presented in Fig. 2. The Mann-Whitney U test revealed that the concentration of ΣNNIs was significantly higher ($P < 0.05$) in sediments in lakes than those in rivers. This probably derives from the fluvial transport to the lake in addition to regional enrichment. The highest concentration of ΣNNIs was detected in Luoma Lake, followed by Old Yellow River, which was the nearest point of access to the Luoma Lake. On the other hand, the greater dilution effect on NNIs by high flow velocity of river was one of the important reasons for the smaller concentrations of NNIs in sediment of the river in comparison to lake (Cheng et al., 2017). The Mann-Whitney U test showed that the pollution degree of NNIs in natural rivers did not differ with artificial rivers (canal) ($P > 0.05$). Therefore, with regard to the enrichment of NNIs in surface sediment might related to river runoff but not of stream formation.

3.2. NNI residues in different regions

To investigate the relationship between NNI concentrations and types of area, locations for collecting surface sediment samples were divided into rural areas, central cities and suburbs (Fig. 3). The arithmetic mean of ΣNNIs followed the order of rural areas (1.94 ng g^{-1} dw), central cities (1.51 ng g^{-1} dw), and suburbs (1.47 ng g^{-1} dw) (from greatest to least). IMIZ, DIN and NIT dominated NNI compositions in three types of regions, which was consistent with the patterns identified in previous studies (Chen et al., 2019a; Chen et al., 2019b). In the rural areas, the heavily polluted area was mainly concentrated in Xuzhou, Suqian and Yancheng City, accounting more than 65% was provided in total NNIs. The regions above were the primary regions of crop farming in Jiangsu Province, such as vegetables (Jiangsu provincial department of agriculture and rural affairs, 2021). Vegetables to be more require for spraying of pesticides than other crops because they are more likely to be susceptible by pests (Li et al., 2014). In the central cities, major NNIs contamination exists in Nanjing, Suzhou, Changzhou and Wuxi City, where had the highest level of economic development and the most urbanized in Jiangsu Province. This phenomenon can originate from the insecticide used in the garden landscape of urban. In addition to the region of protecting crop, urban garden landscape is also one of the major region, where the insecticide has been used in high frequency (Wang et al., 2019). Suburbs is a stage of the process of urbanization in region, urbanized land cover increases and ecological land decreases were their presenting features. The change of land-use type (e.g., farmland converted into asphalt concrete pavement or commercial buildings) might be the main reason given by suburbs with low concentration of NNIs.

3.3. NNI residues in different land use types

To further validate the effect of different land use types around river channels on NNI residues in surface sediment, crop land and construction land are compared in this study. The arithmetic mean of ΣNNIs was 1.84 ng g^{-1} dw in surface sediment of the region around crop land, which was higher than construction land (1.39 ng g^{-1} dw) (Fig. 4). The Mann-Whitney U test showed that the concentration of DIN in surface sediment near crop land was significantly higher than construction land ($P < 0.05$). No significant difference was found among other NNIs at the 0.05 level. The broad applicability of NNIs lessened the discrepancy between different land use types. Previous studies had showed that NNIs were used extensively by pest management on landscaping in urban areas and protect crop in agricultural systems (Bonmatin et al., 2015; Wang et al., 2019). For the DIN, its relative contribution in surface sediment near crop land was the highest (18.76%). Higher point of the residual contamination of NNIs were distributed primarily among the Old Yellow River, Beijing-Hangzhou Canal, Shaobo Lake and Baima Lake, where were dominated by agricultural areas on their upstream. DIN can be used in the cultivation of grape and the fruits on the gardens, which are the dominant crop types in upstream region (Chen et al., 2019a) (Jiangsu provincial department of agriculture and rural affairs, 2021).

3.4. Source apportionment of NNIs

Principle component analysis (PCA) was applied to explore the interrelationships among eight NNIs detected in the surface sediment in Jiangsu Province (Fig. 5), which showed that the cumulative contribution rate of the variance of the first three principal components was 55.9%. In principal component 1 (PC1), CLO, NIT and IMI had higher loading values. CLO was frequently used to against insect pests of corn, rice, orchards, vegetables and ornamentals. NIT was applied to protect rice and glasshouse crops. IMI was employed for lawns, grain crops (e.g., rice and maize) and vegetables (UOH, 2013). In this study, the highest concentration of CLO was found in the region near crop land in

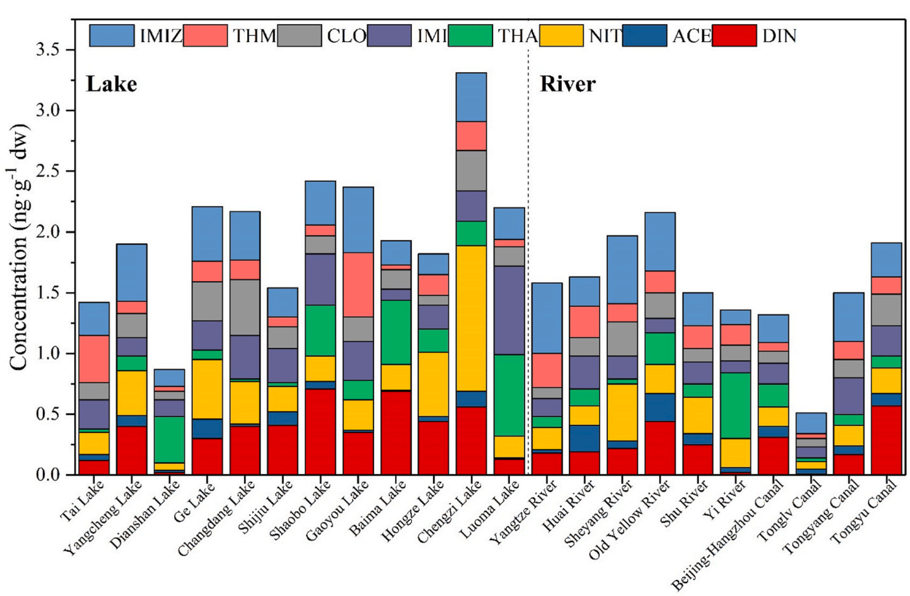

Fig. 2. Total concentrations of NNIs and their relative compositions in surface sediments in Jiangsu Province.

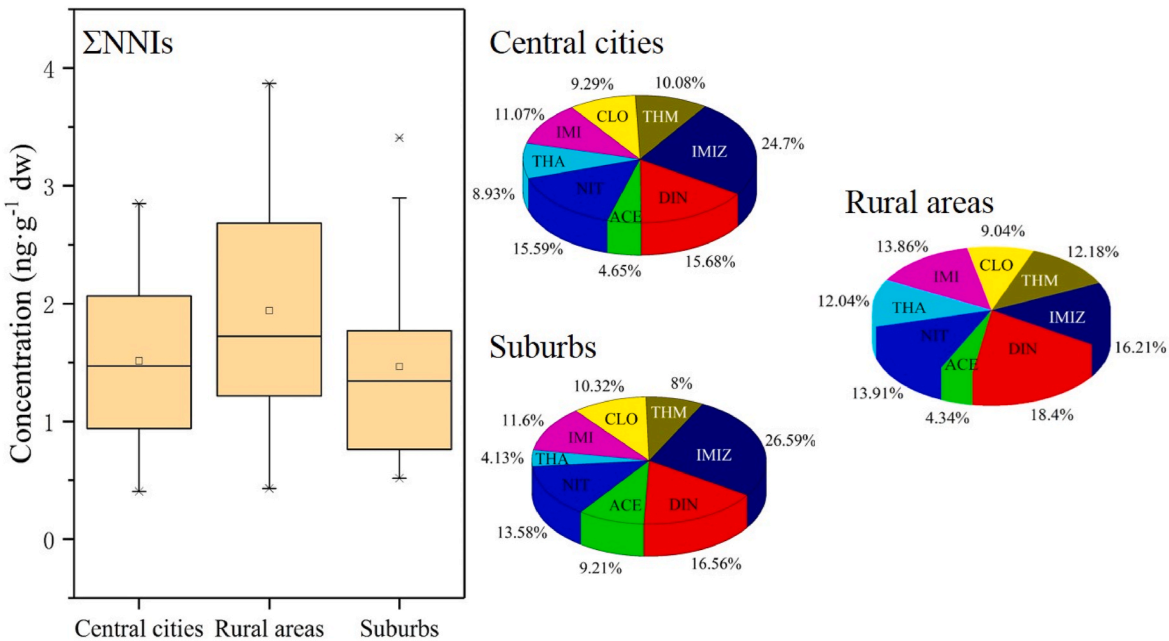

Fig. 3. The concentrations of ΣNNIs and composition profiles of the eight individual NNIs for different types of area: central cities, rural areas and suburbs.

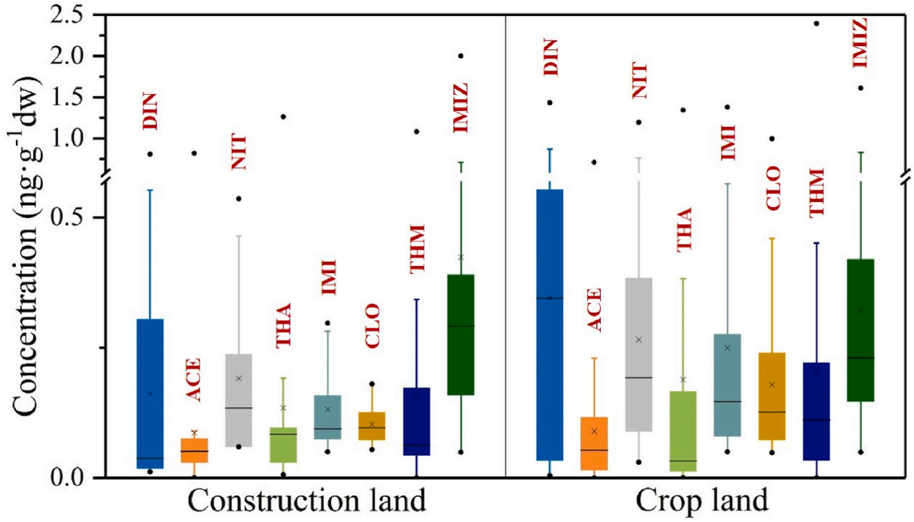

Fig. 4. Boxplots of NNI concentrations in surface sediments in Jiangsu Province. The box ranges from 1st quartile to 3rd quartile, the line in the box is median, the cross mark in the box is the arithmetic mean, the bar represents the whisker range, and the dots outside the box are outliers.

Yancheng City, where was the major producers of rice and pear in Jiangsu Province. The most contaminated stations of NIT and IMI was located in the region downstream of agricultural areas in Xuzhou City, where was the maize and vegetables production main site (Jiangsu provincial department of agriculture and rural affairs, 2021). Combined with regional residual contamination, PC1 is contributed by the pesticide residues in grain crops (maize) and vegetables.

DIN and THA received a high loading score in PC2. DIN mainly originates from the pesticide residues for vegetables, fruit and rice crops, and THA was used to pest control for apples and pears (UOH, 2013). In this study, the highest concentrations of DIN and THA were found to originate from the agricultural areas upstream, where was the major producers of apples, pears and vegetables. Thus, it can be concluded that PC2 coming from crop protection for fruit (apples, pears) and vegetables. ACE was the main payload compound in PC3, which was applied to protect fruit, vegetables and ornamental plants and flowers. The highest concentration of ACE here was found in construction land, indicating the major source of PC3 was landscaping.

3.5. Spatial distribution of NNIs in surface sediment

As for the spatial distribution (Fig. 6), higher concentrations of the majority of NNIs (e.g., DIN, NIT, THA, IMI and IMIZ) were located in the northwest of Jiangsu Province. These were the primary regions of agricultural crops in Jiangsu Province. Under the reported yield statistics of crops, the northwest of Jiangsu Province was the major producers of corn, peanuts, cotton, vegetables, garden fruits, apples, grapes (Jiangsu provincial department of agriculture and rural affairs, 2021). NNIs play an important role in pest control for crops, while residues are more accessible near locale. Notably, as another area with high levels of IMIZ, Nanjing City hold the maximum tea plantation area in Jiangsu Province. The probable reason for this phenomenon is that IMIZ is more

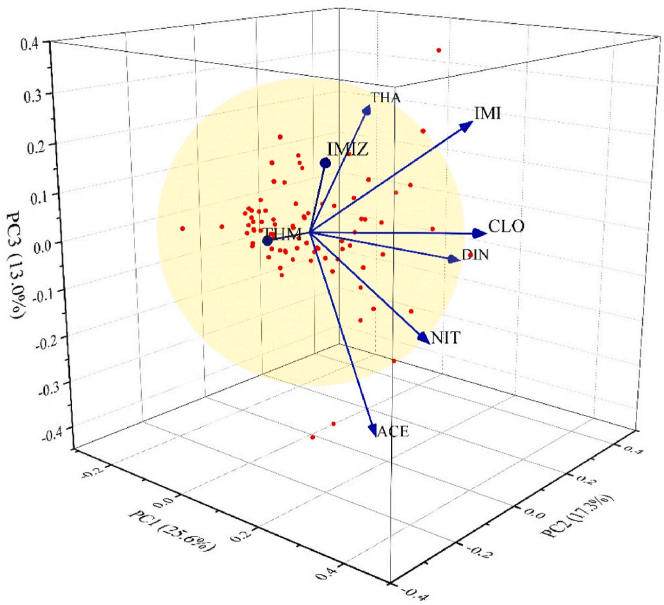

Fig. 5. Plot of principal component analysis (PCA) of NNIs as variables for surface sediments. (Blue arrow means vector resultant of PC1, PC2 and PC3). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

highly amenable to use for tea pest control in comparison with other NNIs. The region with heavier pollution of ACE and CLO was located in northeast (Yancheng City) in Jiangsu Province, where was the major producers of melon fruits. Similarly, this is associated with the applicability of NNIs. In terms of the spatial distribution, the residues of NNIs in surface sediment was predominantly distributed in the northwest and northeast in Jiangsu Province.

4. Conclusion

In summary, we measured the residual concentration of eight individual NNIs in surface sediment in Jiangsu Province, and found NNIs were widespread. Of those, the heavier contaminations of NNI residues were occurred in the northwest and northeast in Jiangsu Province. The distribution of NNIs in surface sediments was determined by the application patterns related to regional characteristics and land use types. Crop types, river runoff and flow rate might be another impact factor for the distribution of NNIs in surface sediment. Principal component analysis showed that residues of NNIs in surface sediment in Jiangsu Province mainly originated from protect crop in agricultural systems.

Author Statement

Chushan Huang: Writing – original draft preparation, Data curation, Formal analysis, Writing- Reviewing and Editing. Pengchong Wen: Investigation, Experimental analysis. Guocheng Hu: Study design, Writing- Reviewing. Juanheng Wang: Assisted in sample collection and examination. Qingyao Wu: Assisted in sample collection and examination. Jianying Qi: Validation and Conceptualization. Ping Ding: Investigation, Data curation. Limei Cai: Investigation, Data curation. Yunjiang Yu: Writing- Reviewing. Lijuan Zhang: Validation and Conceptualization, Writing- Reviewing and Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

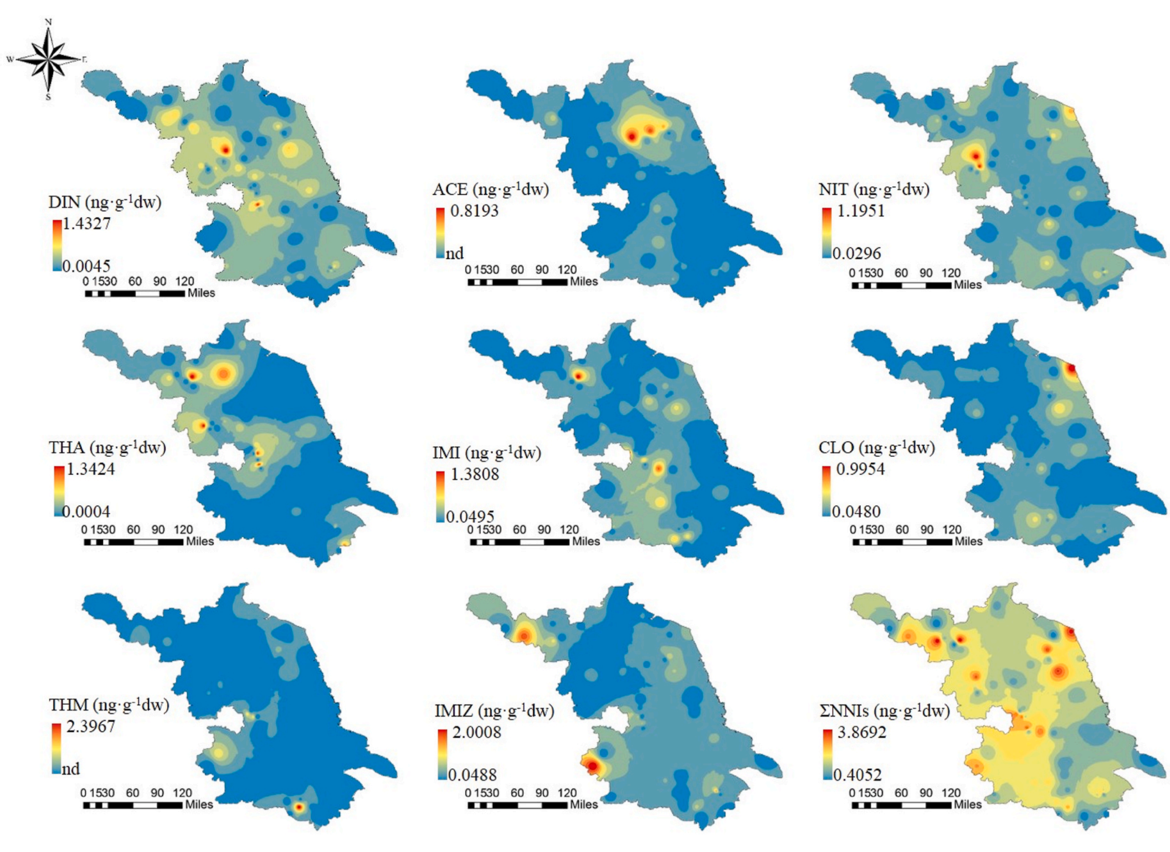

Fig. 6. Spatial distribution of concentrations of NNIs in surface sediments in Jiangsu Province.

Data availability

Data will be made available on request.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2022.120139.

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