Environmental Science and Pollution Research (2023) 30:1039–1047
https://doi.org/10.1007/s11356-022-22270-y

RESEARCH ARTICLE

Meta-analysis of neonicotinoid insecticides in global surface waters

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Received: 28 January 2022 / Accepted: 24 July 2022 / Published online: 30 July 2022

Abstract

Neonicotinoids (NEOs) are a class of insecticides that have high insecticidal activity and are extensively used worldwide. However, increasing evidence suggests their long-term residues in the environment and toxic effects on nontarget organisms. NEO residues are frequently detected in water and consequently have created increasing levels of pollution and pose significant risks to humans. Many studies have focused on NEO concentrations in water; however, few studies have focused on global systematic reviews or meta-analyses of NEO concentrations in water. The purpose of this review is to conduct a meta-analysis on the concentration of NEOs in global waters based on published detections from several countries to extend knowledge on the application of NEOs. In the present study, 43 published papers from 10 countries were indexed for a meta-analysis of the global NEO distribution in water. Most of these studies focus on the intensive agricultural area, such as eastern Asia and North America. The order of mean concentrations is identified as imidacloprid (119.542 ± 15.656 ng L^{-1}) > nitenpyram (88.076 ± 27.144 ng L^{-1}) > thiamethoxam (59.752 ± 9.068 ng L^{-1}) > dinotefuran (31.086 ± 9.275 ng L^{-1}) > imidaclothiz (24.542 ± 2.906 ng L^{-1}) > acetamiprid (23.360 ± 4.015 ng L^{-1}) > thiacloprid (11.493 ± 5.095 ng L^{-1}). Moreover, the relationships between NEO concentrations and some environmental factors are analyzed. NEO concentrations increase with temperature, oxidation–reduction potential, and the percentage of cultivated crops but decrease with stream discharge, pH, dissolved oxygen, and precipitation. NEO concentrations show no significant relations to turbidity and conductivity.

Keywords Neonicotinoids · Insecticide · Organic pollutant · Insecticide contamination · Water · Meta-analysis

Responsible Editor: Ester Heath

Jianqiao Wang and Ru Yin made equal contribution to this work.

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Abbreviations

NEOs	Neonicotinoids
ACE	Acetamiprid
CLO	Clothianidin
DIN	Dinotefuran
IMI	Imidacloprid
IMZ	Imidaclothiz
NIT	Nitenpyram
THI	Thiacloprid
TXM	Thiamethoxam
DO	Dissolved oxygen
ORP	Oxidation-reduction potential
ND	Not detected
NA	Not analyzed
MDL	Method detection limit

Introduction

Neonicotinoids (NEOs) are a class of insecticides that act selectively on nicotinic acetylcholine receptors (nAChRs) to block the action of acetylcholine in the central nervous systems of insects (Matsuda et al. 2001; Tomizawa and Casida 2003). Compared to traditional pesticides, they show stronger selectivity for insects on nAChRs than vertebrates and are thus considered to have reduced toxicity and to exhibit lower resistance in mammals (Jeschke et al. 2013). Since NEOs were first produced in the 1990s beginning with imidacloprid (IMI), other NEOs, including acetamiprid (ACE), clothianidin (CLO), thiamethoxam (TXM), thiacloprid (THI), nitenpyram (NIT), and dinotefuran (DIN), have been successively developed for the market (Godfray et al. 2015). In addition, imidaclothiz (IMZ) is a new NEO with more systemic activity developed by Nantong Jiangshan Agrochemical and Chemical Co. Ltd., China, and it was registered in 2006 by the Chinese Ministry of Agriculture (Shao et al. 2013). NEOs have become best-selling insecticides with annual sales of 1.9 billion dollars, accounting for 25% of the global insecticide market since 2010 (Jeschke et al. 2011). In 2012, TXM, CLO, and IMI accounted for almost 85% of total NEO sales and were mainly used for crop protection (Bass et al. 2015). In particular, IMI has gradually become one of the most widely applied insecticides and is used for over 140 agricultural crops in approximately 120 countries (Drobne et al. 2008). Approximately 20,000 tons of active substance IMI is produced annually, and China contributes approximately 70% of IMI production (Drobne et al. 2008; Simon-Delso et al. 2015; Wang et al. 2018). Because of the highly efficient insect pest control and favorable safety profiles of NEOs, they have been used in agriculture, animal husbandry, and residential environments worldwide (Simon-Delso et al. 2015; Morrissey et al. 2015).

Along with their global use, NEOs have had negative effects on wildlife. Many organisms, including nontarget species and terrestrial pollinators such as bumble bee (*Bombus terrestris*), honey bee (*Apis mellifera*), and butterfly (*Polyommatus icarus*), are extremely sensitive to NEOs (Whitehorn et al. 2012; Rundlöf et al. 2015; Basley and Goulson 2018). Honey bees, as pollinators, play essential roles in ecological systems and crop productivity, so their health, productivity, and behavior are of greater environmental concern (Henry et al. 2012). An increasing number of studies have revealed that NEOs tend to easily enter ecosystems through runoff and drainage systems in agricultural areas and pose increasing ecological threats to organisms (Anderson et al. 2018; Schaafsma et al. 2019). NEOs have the potential to cause a sudden decline in the adult honeybee population, also known as colony collapse disorder (Henry et al. 2012). Many studies have reported on the acute toxicity of NEOs to aquatic invertebrates, birds, and mammals from in vitro and in vivo laboratory toxicity experiments (Morrissey et al. 2015; Han et al. 2018; Addy-Orduna et al. 2019). The potential toxic effects of NEOs mainly include reproductive toxicology, neurotoxicity, hepatotoxicity, immunotoxicity, and genetic toxicity (Han et al. 2018).

Variable levels of NEOs and their metabolites occur in surface environmental media such as soils (Jones et al. 2014; Bonmatin et al. 2019), drinking water (Sultana, et al. 2018), crops (Kamel et al. 2010; Chahil et al. 2015; Karthikeyan et al. 2019), pollen (Tosi et al. 2018), and even bovine milk (Adelantado et al. 2018). It is important to develop better knowledge of the distribution of NEO levels in the environment and the associated environmental effects, which will help guide conservation efforts to NEOs application and environment protection. Meta-analysis is a quantitative method to summarize the independent research results. Hence, the objective of this review is to summarize the global concentration distribution of NEOs (ACE, CLO, DIN, IMI, IMZ, NIT, THI, and TXM) in water and reveal the relationship between NEO concentrations and hydrologic parameters such as stream discharge, turbidity, pH, temperature, dissolved oxygen (DO), oxidation–reduction potential (ORP), precipitation, and cultivated crops via meta-analysis.

Materials and methods

Data assembly

To study NEO levels in water, target publications included in the PubMed database were screened on February 2, 2021. A total of 57 papers were obtained using the following search terms: (((neonicotinoid[Title]) OR (neonicotinoids[Title]) OR (neonicotinoid insecticide[Title]) OR (neonicotinoid insecticides[Title])) AND ((water[Title]) OR (lake[Title]) OR (river[Title]) OR (stream[Title]) OR (wetland[Title]))). Among the papers obtained, 27 were retained in the present study based on the following criteria: (1) papers written in English were retained; (2) duplicate papers were removed; (3) irrelevant papers were carefully removed after reading the abstracts; (4) papers excluding NEO concentration data were removed after reading the full text in detail; and (5) papers were identified as original research rather than review articles. An additional 16 papers were obtained from the references of the retained papers, so a total of 43 papers were used in this study. These selected papers were published from 2012 to 2021 with the impact factor range from 1.755 to 11.236. Although they might be not comprehensive, the papers that we screened were published in specialized journals with considerable impact. The following information was extracted: sampling time, country, sampling location, physical and chemical properties of the studied water (stream discharge, turbidity, pH, temperature, DO, ORP, and conductivity), precipitation, percentage of cultivated crops, types of NEOs, concentrations of NEOs (maximum, median, minimum, and mean), and standard deviation of NEO concentrations. These studies referring to 10 countries (the USA, Australia, Belize, Canada, China, Japan, the Philippines, Romania, South Africa, and

Vietnam) were selected. NEOs were detected in tap water, seawater, lakes, rivers, reservoirs, estuaries, creeks, wetlands, or open ditches and runoff in agricultural regions whether it's spring, summer, fall or winter (Table S1). Plot Digitizer software was used to extract values from graphs.

Data analysis

The sampling locations were displaced on a world map based on longitude and latitude parameters by RStudio (Fig. 1). With no information on longitude and latitude, the sampling site name was used to extract longitude and latitude information from Google Maps. The mean concentration of each NEO was used, and the concentrations of NEOs were unified to ng L^{-1} for further analysis. Data analyses and the meta-analysis figures were developed using the JMP statistical program (version 16.0). JMP is a statistical visualization tool, it can integrate the graphics into the report. The "Distribution of Y" platform was used for testing the mean concentrations of different NEOs. The number of observations and concentration range for different NEOs (ACE, CLO, DIN, IMI, IMZ, NIT, THI, and TXM) were summarized. The "Fit Y by X" platform was used for testing the significant differences between the mean concentrations of NEOs and environmental factors (e.g., stream discharge, turbidity, pH, temperature, DO, ORP, conductivity, precipitation, and the percentage of cultivated crops).

Results and discussion

Database availability

The main regions exhibiting NEO use in agriculture are 29.4% of total global use in Latin America, followed by 23% in Asia, North 22% in America, and 11% in Europe (Bass et al. 2015; Simon-Delso et al. 2015). Most of our selected studies focus on eastern Asia and North America, which include countries heavily focused on agricultural production (Fig. 1). However, no study about Latin America was obtained in the present study. The mean concentrations of eight widely used NEOs (ACE, CLO, DIN, IMI, IMZ, NIT, THI, and TXM) were collected, and the information on each form of NEO detected is shown in Fig. 2. IMI is the most frequently reported (39/43, 91%), followed by CLO (36/43, 84%), TXM (32/43, 74%), ACE (31/43, 72%), THI (27/43, 63%), DIN (16/43, 37%), NIT (11/43, 26%), and IMZ (4/43, 9%). IMI, the first NEO developed, is the most frequently reported, possibly due to its broad application and usage (Kollmeyer et al. 1999). IMZ was the latest to enter the market; thus, there only a few studies include IMZ detection. Continuous detection of IMZ in the environment is necessary, because it has great potential in China's market.

NEO concentrations in water

Table 1 shows the concentrations and numbers of observations for different NEOs. CLO was the most frequently detected in 1056 out of 1645 water samples, followed by IMI (879), TXM (863), ACE (428), THI (295), DIN (122), IMZ (37), and NIT (29). CLO has the highest mean concentrations at 222.320 ± 46.692 ng L^{-1}. The mean concentrations of other NEOs are ordered as follows: IMI (119.542 ± 15.656 ng L^{-1}) > NIT (88.076 ± 27.144 ng L^{-1}) > TXM (59.752 ± 9.068 ng L^{-1}) > DIN (31.086 ± 9.275 ng L^{-1}) > IMZ (24.542 ± 2.906 ng L^{-1}) > ACE (23.360 ± 4.015 ng L^{-1}) > THI (11.493 ± 5.095 ng L^{-1}). Moreover, concentrations were found to range from 0.001 to 45,100 ng L^{-1} for CLO, from

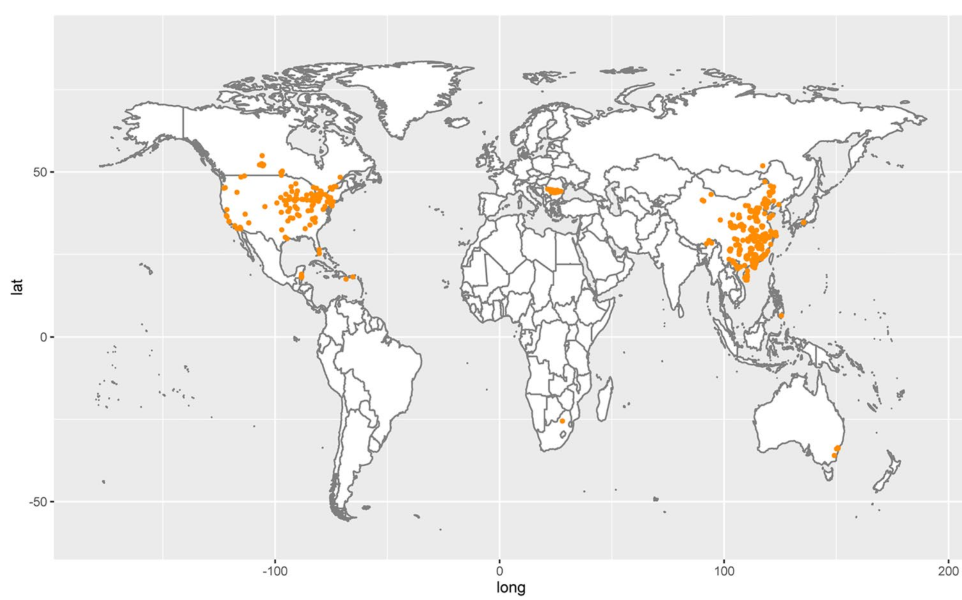

Fig. 1 Geographic focuses of field studies investigating concentrations of NEOs in water worldwide

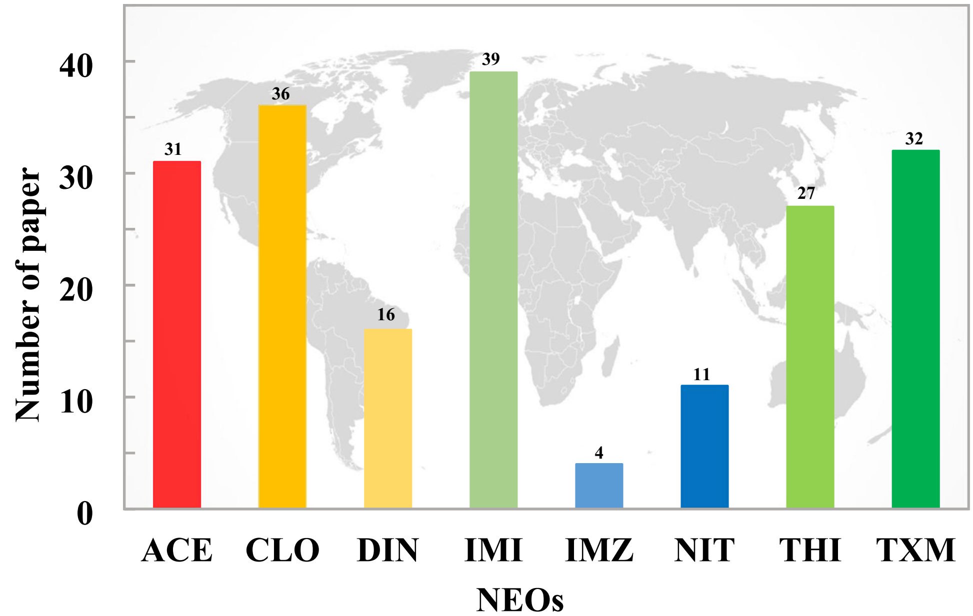

Fig. 2 Number of papers focused on each NEO concentration in water

Table 1 Summary of the dataset indicating the number of observations for different NEO types (ACE, CLO, DIN, IMI, IMZ, NIT, THI, TXM), and statistics (mean ± standard error (SE), lower 95% confidence interval (LCI), upper 95% confidence interval (UCI)), and the ranges of concentrations of each NEO type

Type	*n*	Mean (ng L^{-1})	SE	Range (ng L^{-1})	LCI	UCI
ACE	428	23.360	4.015	(0.0025, 1527.6)	15.469	31.252
CLO	1056	222.320	46.692	(0.001, 45,100)	130.700	313.939
DIN	122	31.086	9.275	(0.11, 1022.2)	12.725	49.448
IMI	879	119.542	15.656	(0.004, 9140)	88.813	150.270
IMZ	37	24.542	2.906	(0.002, 81.92)	18.648	30.436
NIT	29	88.076	27.144	(2, 672.9)	32.475	143.678
THI	295	11.493	5.095	(0.003, 1370)	1.466	21.520
TXM	863	59.752	9.068	(0.002, 3820)	41.960	77.543

0.004 to 9140 ng L^{-1} for IMI, from 0.002 to 4315 ng L^{-1} for TXM, from 0.002 to 3820 ng L^{-1} for ACE, from 0.003 to 1370 ng L^{-1} for THI, from 0.11 to 1022.2 ng L^{-1} for DIN, from 2 to 672.9 ng L^{-1} for NIT, and from 0.002 to 81.92 ng L^{-1} for IMZ (Table 1).

Figure 3 displays the distributions of the mean concentrations of each NEO type. The concentrations of CLO and IMI were found to be concentrated at 0 ~ 1500 ng L^{-1} and 0 ~ 500 ng L^{-1}, respectively. The concentrations of ACE, DIN, IMZ, NIT, THI, and TXM were mainly measured at below 250 ng L^{-1}. NEOs can be used in pest control to protect crops and are mainly applied for seed treatment, chemigation, and soil treatment (Simon-Delso et al. 2015). NEOs may enter through various media into aquatic systems from agricultural fields through processes such as spray drift, atmospheric deposition, soil erosion, and runoff. Some governments and organizations have established water quality guidelines for protecting aquatic ecosystems. For example, the US Environmental Protection Agency (USEPA) has estimated that chronic benchmarks of 970, 2100, 10, 740, 95, 300, and 50 ng L^{-1} for THI, ACE, IMI, TXM, DIN, and CLO, respectively (USEPA 2021). In this review, some potentially threatening concentrations of certain NEOs are especially found in agricultural regions. THI monitored at the outlet of the Yarramundi Lagoon in a turf farm was found the highest concentration of 1370 ng L^{-1} (Sánchez-Bayo and Hyne 2014). The highest IMI concentration found in Solomon Creek in the Californian agricultural region was recorded as 9140 ng L^{-1} (Anderson et al. 2018). Although the province of Ontario of Canada bans the cosmetic use of some pesticides on lawns and gardens, NEOs are used for seed treatment on row crops such as corn, soybeans, cereal grains, and canola, which has led to widespread use in Ontario (Ontario Class 9 pesticides 2016). CLO, TXM, and ACE levels in drain water around maize fields in Canada have reached 45,100 and 7200, 4315, and 1527.6 ng L^{-1}, respectively (Schaafsma et al. 2019).

In recent years, the European Union has banned some NEOs because of their improvement in the decline of bees and other pollinators (Naumann et al. 2022). However,

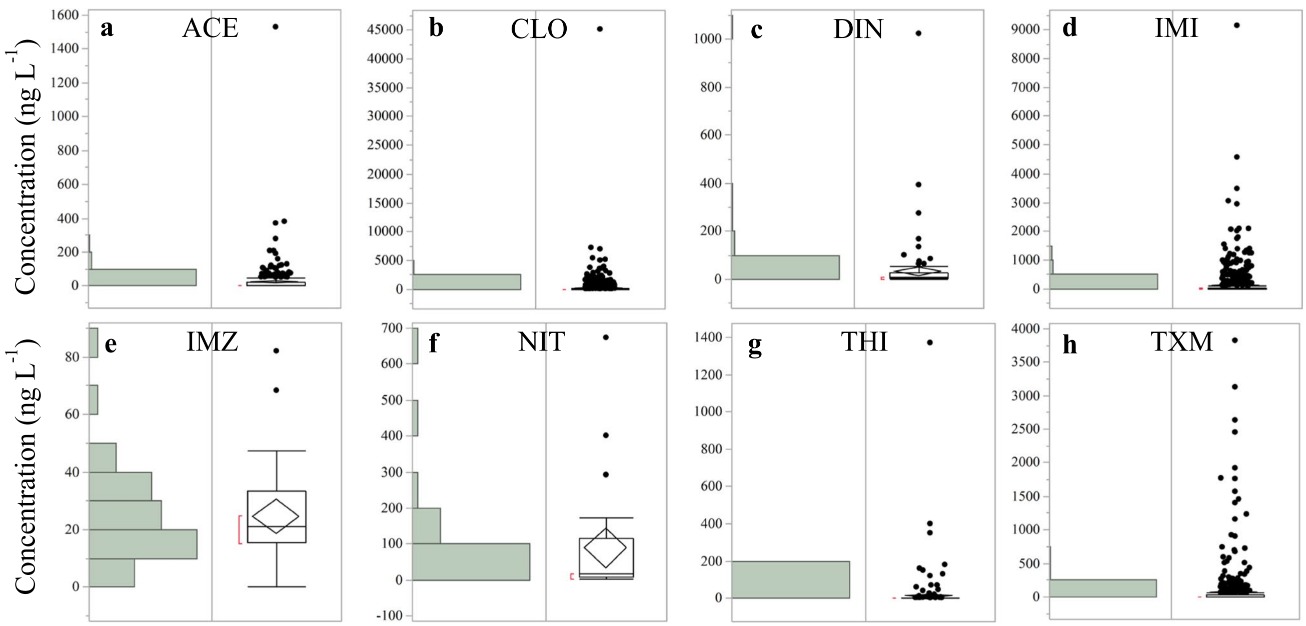

Fig. 3 Distribution of mean concentrations of each NEO (**a** ACE; **b** CLO; **c** DIN; **d** IMI; **e** IMZ; **f** NIT; **g** THI; **h** TXM). The top and bottom of the diamond (graph on the right) are a 95% confidence interval for the mean. The bottom and top of the box show the 25th and 75th quantiles, and median is the horizontal line inside the box

NEOs are still widely used in developing countries with poorly controlled. China has the highest production of NEOs, which are frequently detected in rivers flowing through urban environments. In addition to those found in agricultural regions, the highest concentrations of DIN, NIT, and IMZ have been detected in the Yangtze River in China, reaching levels of 1022.3, 672.9, and 81.92 ng L^{-1}, respectively (Chen et al. 2019). The Yangtze River is the longest river in China, playing a considerable role in agricultural and industrial activities (Mahai et al. 2019). NEOs in the Yangtze River have become a source of NEOs in seawater (Chen et al. 2019). Although NEO concentrations decrease rapidly by dilution, NEOs are detected near shorelines (Pan et al. 2020). IMZ is a novel NEO that has been gradually applied to vegetables, fruits, and crops on a large scale in China because of its excellent insecticidal activity (Tao et al. 2021). Due to IMZ's increasing use, more attention should be dedicated to its adverse effects (e.g., DNA damage in earthworms; Zhang et al. 2017). Moreover, different NEO concentrations have been detected in different crop planting periods. Concentrations of IMI and TXM increase markedly in the rice planting month. DIN was detected at a concentration of 220 ng L^{-1} during rice earwig emergence (Yamamoto et al. 2012). A large proportion of pesticides enter environmental media via runoff, leaching, and drifting. These pesticides are absorbed by nontarget plants or organisms and present a potential threat to food safety (Li et al. 2018; Tao et al. 2021). Thus, scientists around the world have gradually recognized NEO risks and increased efforts to monitor NEOs in the environment (Morrissey et al. 2015).

Effect of physicochemical properties on NEO concentration

Figure 4 and Table 2 present the relationship between NEO concentrations and nine physical and chemical properties. Different properties show different responses to NEO concentrations in water. NEO concentrations increase with temperature, ORP, and the percentage of cultivated crops (line regression, temperature: adjusted $R^2 = 0.0811$, $p < 0.0001$; ORP: adjusted $R^2 = 0.0931$, $p < 0.01$; cultivated crop: adjusted $R^2 = 0.0307$, $p < 0.001$) (Fig. 4d, f, and i). When summer arrives, pest damage increases with increasing temperature, and insecticide use is increased to decrease crop losses. Rainfall is a key factor in increasing NEO residues in water. NEOs can enter water via surface and underground runoff, creating higher insecticide concentrations in water. For instance, in the province of Guangdong located in the subtropical zone of South China, the climate is warm and humid for most of the year. Thus, large quantities of pesticides are used for pest control, and Guangdong Province has the highest pesticide application dosage (Li et al. 2014). Only one paper presents the value of ORP, and the representativeness of the relation needs to be further confirmed (Yi et al. 2019). Concentrations of NEOs generally increase as the percentage of cultivated crops increases. High NEO concentrations are detected in surface water around areas

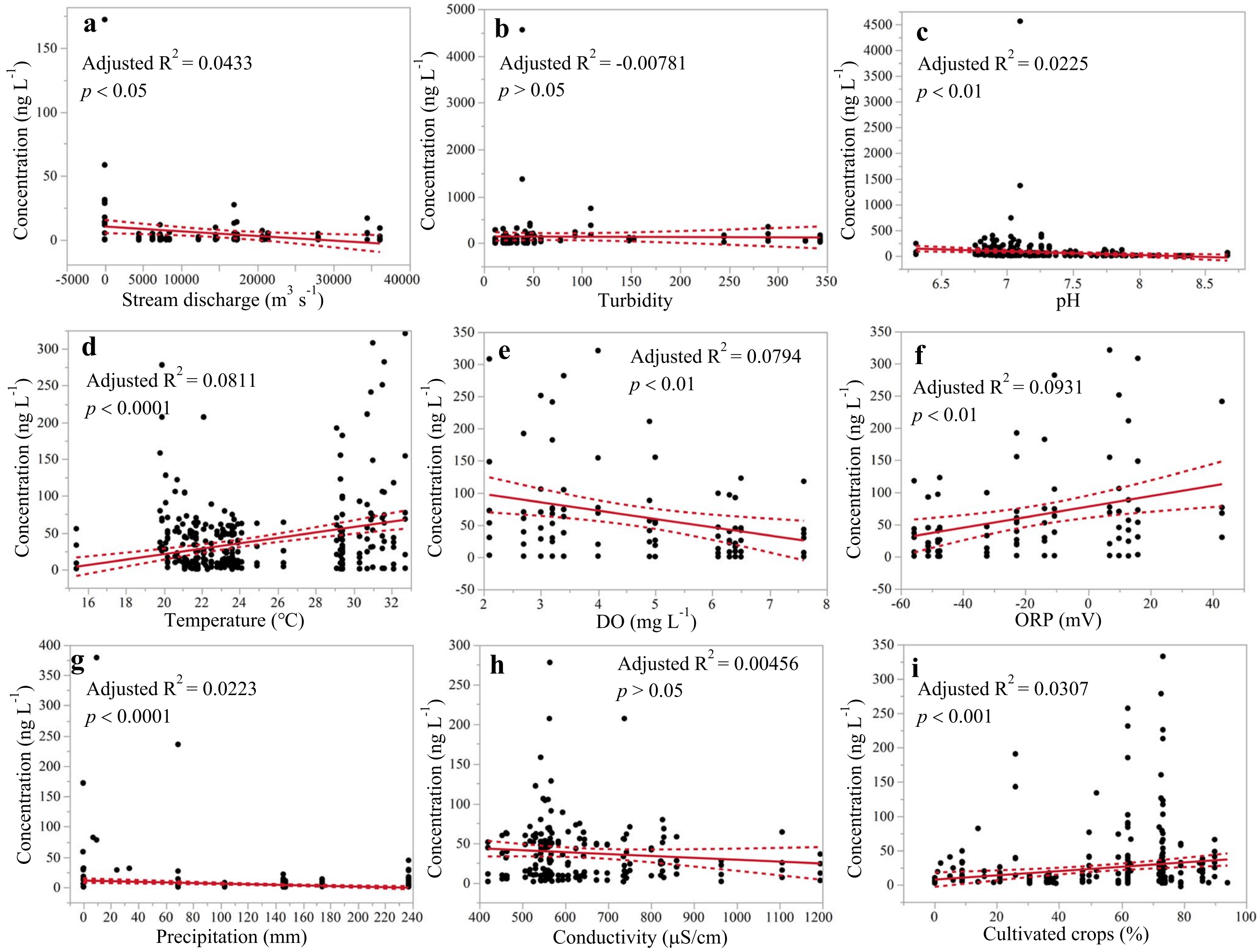

Fig. 4 NEO concentration responses to the effects of stream discharge (**a**), turbidity (**b**), pH (**c**), temperature (**d**), DO (**e**), ORP (**f**), precipitation (**g**), conductivity (**h**), and the percentage of cultivated crops (**i**). $p<0.05$, statistically significant change; $p<0.001$, highly statistically significant

Table 2 Description of the models that explain the relationships between mean concentrations of NEOs and stream discharge, turbidity, pH, temperature, dissolved oxygen, ORP, precipitation, conductivity, and the percentage of cultivated crops

Model	R^2	Adjusted R^2	*F*-value	*p*	*n*
Mean concentration = 10.545 − 0.000368 × stream discharge	0.0510	0.0433	$F_{1,125}=6.658$	0.011	126
Mean concentration = 141.816 − 0.0639 × turbidity	0.000187	−0.00781	$F_{1,126}=0.0234$	0.879	127
Mean concentration = 607.822 − 73.932 × pH	0.0248	0.0225	$F_{1,429}=10.872$	0.0011	430
Mean concentration = − 53.602 + 3.708 × temperature	0.0839	0.0811	$F_{1,339}=30.954$	< 0.0001	340
Mean concentration = 124.006 − 12.910 × DO	0.0906	0.0794	$F_{1,82}=8.0743$	0.0057	83
Mean concentration = 77.593 + 0.817 × ORP	0.104	0.0931	$F_{1,82}=9.421$	0.0029	83
Mean concentration = 10.796 − 0.0497 × precipitation	0.0236	0.0223	$F_{1,734}=17.736$	< 0.0001	735
Mean concentration = 52.817 − 0.024 × conductivity	0.0104	0.00456	$F_{1,170}=1.778$	0.184	171
Mean concentration = 7.237 + 0.314 × cultivated crops (%)	0.0336	0.0307	$F_{1,331}=11.480$	0.0008	332

of agricultural activity when the planting season arrives. According to a study conducted in the USA, streams show higher NEO concentrations in the planting season than in other seasons (Hladik and Kolpin 2016). Another study from Canada shows that one side of the Two Mile Creek watershed includes over 50% orchards, and an IMI concentration of 816 ng L^{-1} was detected in this creek (Struger et al. 2017). A positive relationship between cultivated crops and NEO concentrations has been observed in other studies (Hladik et al. 2014; Iancu et al. 2019).

NEO concentrations decrease with stream discharge, pH, DO, and precipitation (line regression, stream discharge: adjusted $R^2 = 0.0433$, $p > 0.05$; pH: adjusted $R^2 = 0.0225$, $p < 0.01$; DO: adjusted $R^2 = 0.0794$, $p < 0.01$; precipitation: adjusted $R^2 = 0.0223$, $p < 0.0001$) (Fig. 4a, c, e, and g). The negative relation between NEO concentrations and stream discharge or precipitation may be caused by the dilution of NEOs when strong precipitation occurs (Struger et al. 2017). Higher DO value of water might affect the degradation of NEOs (Yi et al. 2019). The pH value is an important factor that affects NEO solubility in water. NEOs have longer term residuals under acidic, or neutral conditions than under less alkaline conditions (Yi et al. 2019). It was reported that NEOs hardly degrade at pH 4.0 ~ 7.0, while NEOs hydrolyze readily with a high pH value (pH = 10). (Todey et al. 2018). In this review, pH values of water samples were ranged from 6.31 to 8.67, suggesting that NEOs might be presented in waters for a long time.

The NEO concentrations show no significant correlations with turbidity, and conductivity ($p > 0.05$) (turbidity: adjusted $R^2 = -0.00781$, $p = 0.879$; conductivity: adjusted $R^2 = 0.00456$, $p = 0.184$) (Fig. 4b and h). NEOs are more likely to dissolve than combine with particulate, or colloidal matter (Sánchez-Bayo and Hyne 2014). However, these relationships need further confirmation.

Conclusions and avenues for future research

In the present work, we summarize a total of 43 publications on NEOs detected in tap water, seawater, lakes, rivers, reservoirs, estuaries, creeks, wetlands, open ditches, and runoff in agricultural regions worldwide. Most studies have focused on eastern Asia and North America, which are major areas of agricultural production. The order of reporting frequency is IMI > CLO > TXM > ACE > THI > DIN > NIT > IMZ. Underdeveloped areas such as Africa should be considered due to an increasing use of NEOs in these areas. In addition, the order of mean concentrations is IMI > NIT > TXM > DIN > IMZ > ACE > THI. The highest IMI concentration (9140 ng L^{-1}) was detected in Solomon Creek in the Californian agricultural region of the USA, while THI (1370 ng L^{-1}) was monitored at the outlet of the Yarramundi Lagoon in Australia. The highest concentrations of CLO (45,100 ng L^{-1}, 7200 ng L^{-1}), TXM (4315 ng L^{-1}), and ACE (1527.6 ng L^{-1}) were found in drain water around maize fields in Canada, and DIN (1022.3 ng L^{-1}), NIT (672.9 ng L^{-1}), and IMZ (81.92 ng L^{-1}) were detected in the Yangtze River in China. Moreover, the relationships between mean concentrations of NEOs and environmental factors (e.g., stream discharge, turbidity, pH, temperature, DO, ORP, conductivity, precipitation, and the percentage of cultivated crops) show that NEO concentrations increase with temperature, oxidation–reduction potential, and the percentage of cultivated crops but decrease with stream discharge, pH, DO, and precipitation. NEO concentrations have no significant relationship to turbidity, and conductivity. To prevent NEO pollution, NEO levels in the environment should be constantly monitored and evaluated.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s11356-022-22270-y.

Author contribution JW and RY contributed to the study conceptualization and design. Investigation, data curation, and formal analysis were performed by JW, RY, YL, and BW. The original draft of the manuscript was written by JW and RY and the revision and editing of the manuscript was commented by NW, PX, TX, and HH. All authors read and approved the final manuscript.

Funding This study was funded by the National Natural Science Foundation of China (grant number 42007311), Natural Science Foundation of Guangdong Province (grant number 2020A1515110189), and the Basic Research Program of Guangzhou, China (grant number 202002030090, 202102010493).

Data availability All data generated or analyzed during this study are included in this article and its supplementary information Table S1.

Declarations

Ethical approval There are no ethical issues in this article.

Consent to participate All the authors agree to participate in this paper.

Consent for publication All the authors agree to publish this paper.

Competing interests The authors declare no competing interests.

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