Meta-Analysis of Neonicotinoid Insecticides in Global Surface Waters

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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 residual 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 conducted surveys of NEO concentrations in water; however, few studies have focused on global systematic reviews or meta-analyses of NEO concentrations in water. In the present study, 43 published papers from 10 countries were indexed for a meta-analysis of the global NEO distribution in water. Among these studies, most focus on eastern Asia and North America, which are involved in intensive agricultural activities. 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 relationship between NEO concentrations and some environmental factors is 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. 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.

1. 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 to mammals. 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. 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 humble bees, honey bees and butterflies, 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. 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, which is called 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, drinking water, crops, pollen, and even bovine milk (Kamel et al. 2010; Jones et al. 2014; Chahil et al. 2015; Adelantado et al. 2018; Tosi et al. 2018; Sultana, et al. 2018; Bonmatin et al. 2019; Karthikeyan et al. 2019). It is important to develop better knowledge of the distribution of NEO levels in the environment and the associated environmental effects. 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.

2. Materials And Methods

2.1 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) duplicate papers were removed; (2) irrelevant papers were carefully removed after reading the abstracts; (3) papers excluding NEO concentration data were removed after reading the full text in detail; and (4) papers were identified as original research rather than review articles. Then, 16 papers were obtained from the related references of the above selected papers. Finally, 43 papers were used in this study. Although they might be not comprehensive, the papers that we screened were published in specialized journals with considerable impact. The following information was extracted: the sampling time, the country, the sampling location, physical and chemical properties of the studied water (stream discharge, turbidity, pH, temperature, DO, ORP, and conductivity), precipitation, the percentage of cultivated crops, types of NEOs, concentrations of NEOs (max, median, min and mean concentrations), and the standard deviation of NEO concentrations. When data were displayed in graphs, Plot digitizer software was used to extract values.

2.2 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). The number of observations and concentration range for different NEOs (ACE, CLO, DIN, IMI, IMZ, NIT, THI and TXM) were summarized. A one-way analysis of variance (ANOVA) was used to test 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).

3. Results And Discussion

3.1 Database availability

A total of 43 studies referring to 10 counties (the United States, Australia, Belize, Canada, China, Japan, the Philippines, Romania, South Africa and Vietnam) were selected, and in these studies NEOs were detected in tap water, seawater, lakes, rivers, reservoirs, estuaries, creeks, wetlands, or open ditches and runoff in agricultural regions (Table S1). Most of the studies focus on eastern Asia and North America, which include countries heavily focused on agricultural production (Fig. 1). The main regions exhibiting NEO use in agriculture are Latin America (29.4%), Asia (23%), North America (22%), and Europe (11% of total global use) (Bass et al. 2015; Simon-Delso et al. 2015). 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 detection 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). Only a few studies include IMZ detection, but this should not be ignored. NEO pollution in some underdeveloped areas, such as Africa, should be considered.

3.2 NEO concentrations in water

Table 1 shows the concentrations and numbers of observations for different NEOs. CLO was the most frequently detected in 1056 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 45100 ng L^{-1} for CLO, from 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).

Table 1
Summary of the dataset indicating the number of observations for different NEO types (ACE, CLO, DIN, IMI, DIN, 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, 45100]	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

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}. However, some unreasonably elevated concentrations of certain NEOs are especially found in agricultural regions. 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. THI monitored at the outlet of the Yarramundi Lagoon in a turf farm was found to reach levels of up to 1370 ng L^{-1} (Sánchez-Bayo et al. 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 (Octario 2016). CLO, TXM and ACE levels in drain water around maize fields in Canada have reached 45100 and 7200, 4315, and 1527.6 ng L^{-1}, respectively (Schaafsma et al. 2019). 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. NEOs in the Yangtze River have become a source of NEOs in seawater. 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). It has been reported that IMZ can cause DNA damage in earthworms (Zhang et al. 2017). Due to IMZ's increasing use and adverse effects on nontargeted organisms, increasing attention should be dedicated to this NEO. 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.

3.3 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 = 0.0029; Cultivated crop: adjusted R^2 = 0.0307, p = 0.0008) (Fig. 4d, f, 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 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).

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

NEO concentrations decrease with stream discharge, pH, DO and precipitation (Line regression, Stream discharge: adjusted R^2=0.0433, p=0.011; pH: adjusted R^2 = 0.0225, p = 0.0011; DO: adjusted R^2 = 0.0794, p = 0.0057; Precipitation: adjusted R^2 = 0.0223, p < 0.0001) (Fig. 4a, c, e, 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). 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). The self-purifying ability of a water body has a great effect on the degradation of NEOs, following higher DO concentrations.

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, 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.

4. 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 (45100 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.

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.

Declarations

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Availability of data and materials: All data generated or analyzed during this study are included in this article and its supplementary information Table S1.

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

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Figures

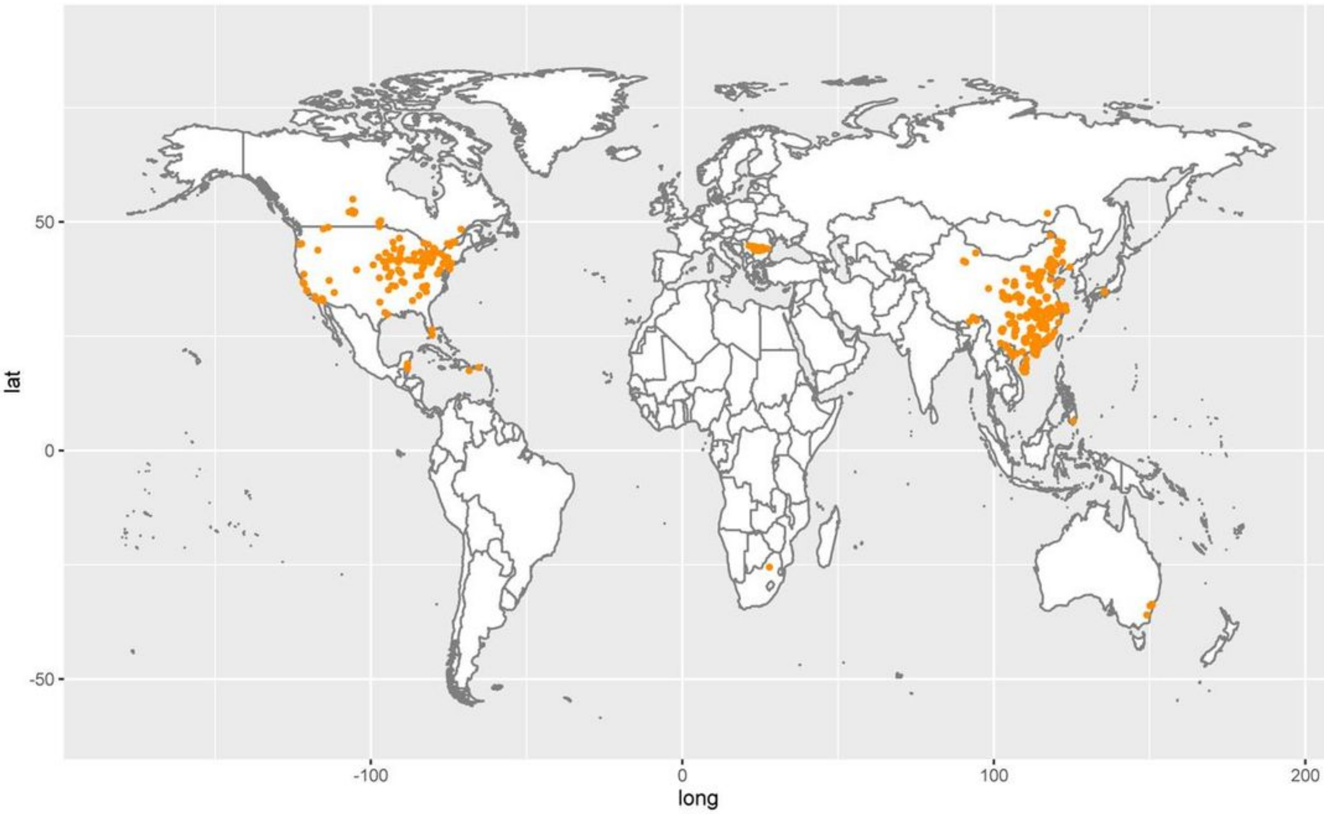

Figure 1

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

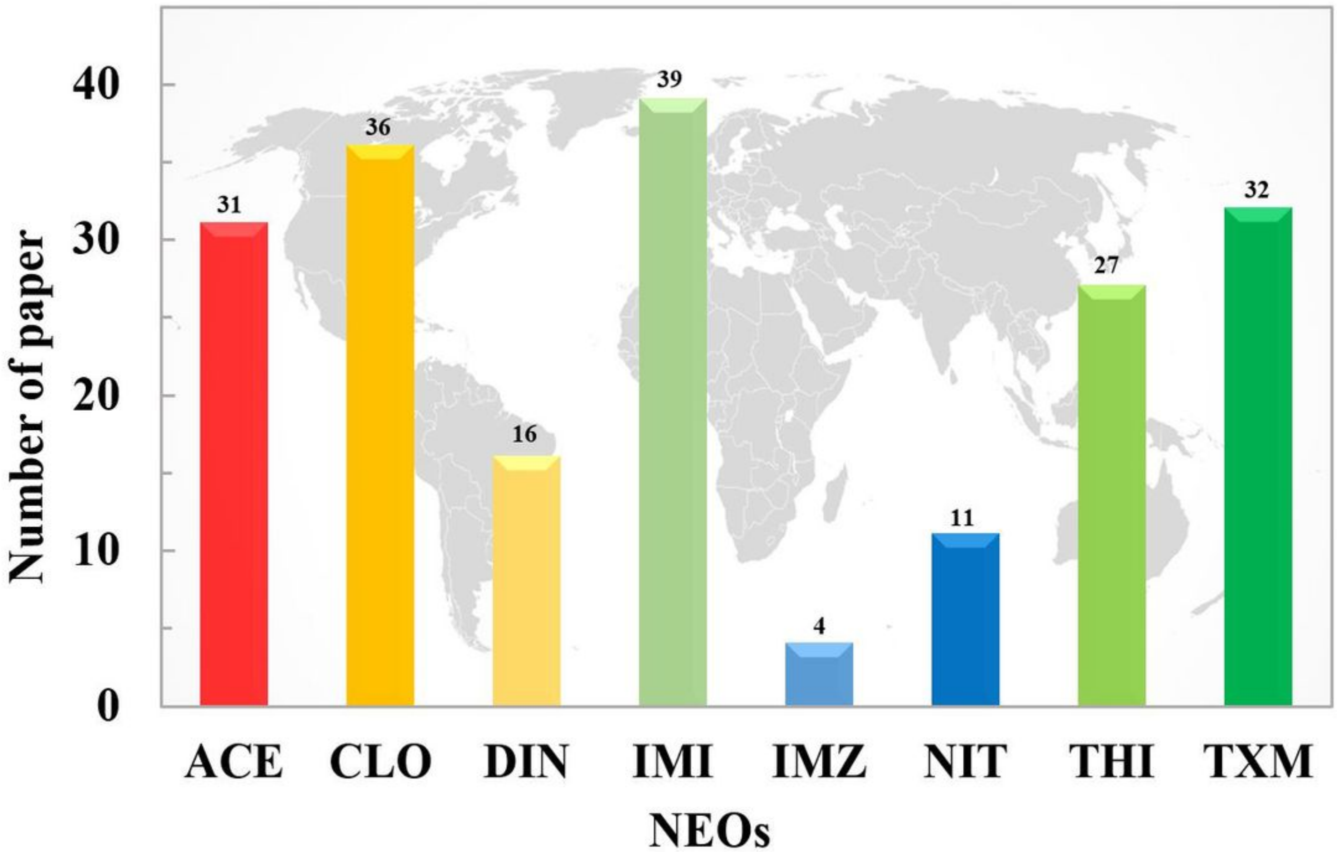

Figure 2

Number of papers focused on each NEO concentration in water.

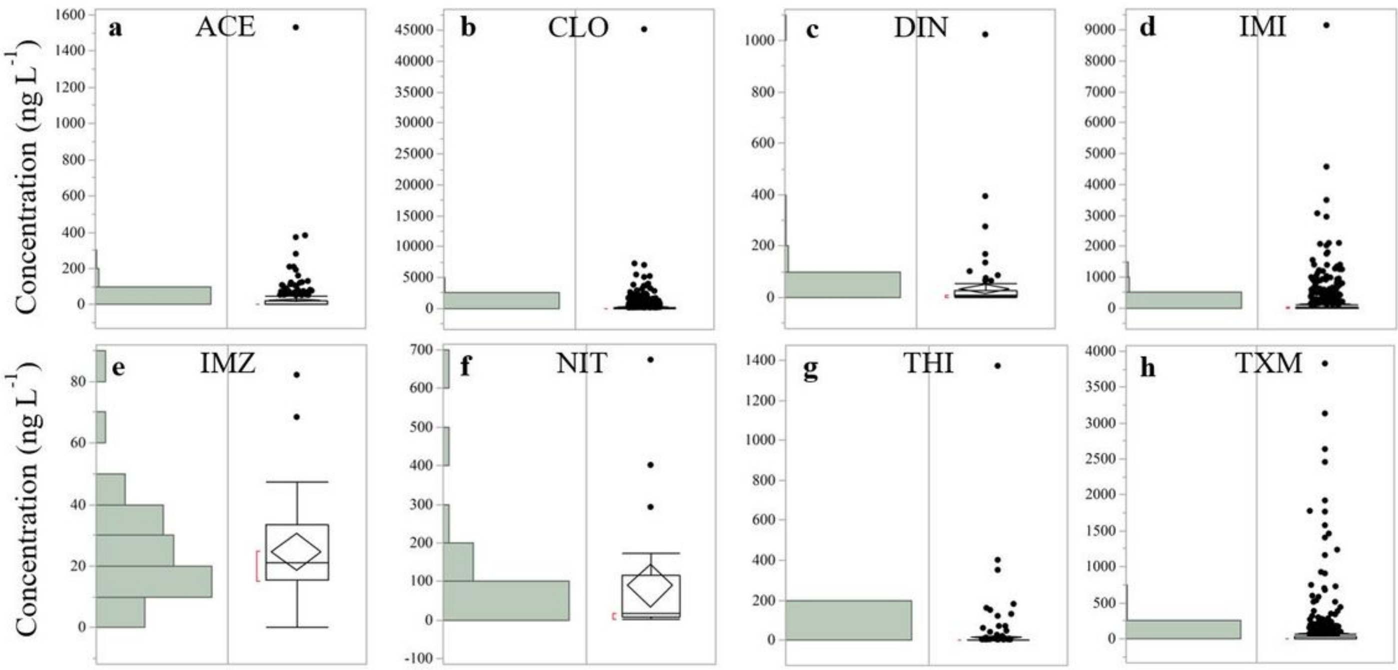

Figure 3

Distribution of mean concentrations of each NEO (a: ACE; b: CLO; c: DIN; d: IMI; e: IMZ.; f: NIT; g: THI; h: TXM)

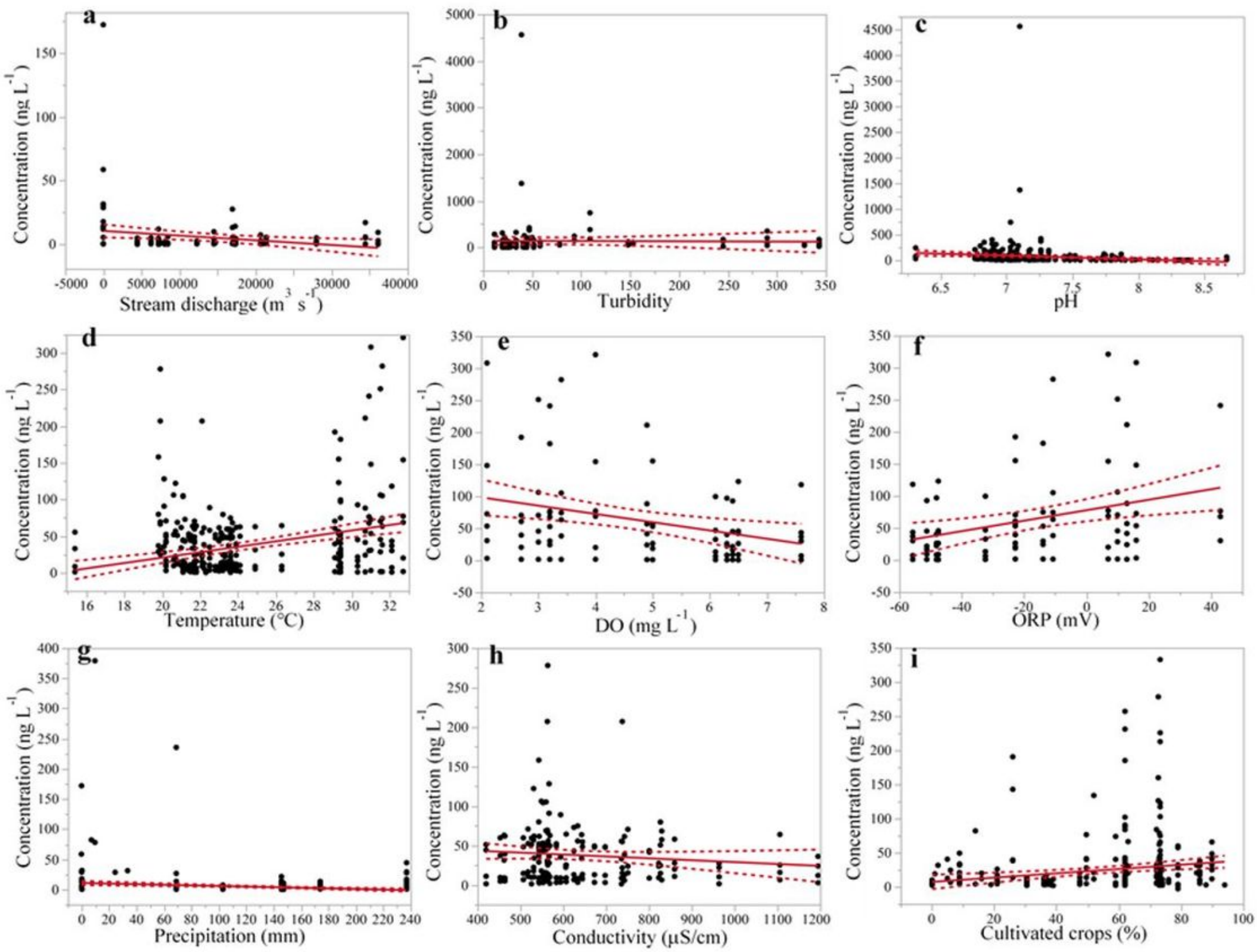

Figure 4

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).

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- TableS1SupplementarydataRawdata.xlsx