REPORT

NEONICOTINOIDS

Neonicotinoids disrupt aquatic food webs and decrease fishery yields

Masumi Yamamuro[1,2]*, Takashi Komuro[2], Hiroshi Kamiya[3], Toshikuni Kato[3], Hitomi Hasegawa[4], Yutaka Kameda[5]

Invertebrate declines are widespread in terrestrial ecosystems, and pesticide use is often cited as a causal factor. Here, we report that aquatic systems are threatened by the high toxicity and persistence of neonicotinoid insecticides. These effects cascade to higher trophic levels by altering food web structure and dynamics, affecting higher-level consumers. Using data on zooplankton, water quality, and annual fishery yields of eel and smelt, we show that neonicotinoid application to watersheds since 1993 coincided with an 83% decrease in average zooplankton biomass in spring, causing the smelt harvest to collapse from 240 to 22 tons in Lake Shinji, Shimane Prefecture, Japan. This disruption likely also occurs elsewhere, as neonicotinoids are currently the most widely used class of insecticides globally.

Lacustrine ecosystems worldwide yield hundreds of tons of fish annually (*1*); however, in Japan, these ecosystems are being severely degraded by human activities (*2*). The introduction of exotic freshwater piscivorous fishes is reported as a primary cause for reduced fish yields (*2*); however, this impact is negligible because invasive freshwater fishes are scarce in half of the 23 brackish lakes in Japan experiencing declining fish yields. Adverse effects of pesticides are also a likely driver of the reduction in fish yields in Japan, but evidence for this has been lacking until now.

Neonicotinoids are used heavily in rice agriculture (*3*) and were first registered in Japan in 1992. Since then, their use has increased exponentially (Fig. 1). Currently, even rivers in metropolitan areas are contaminated with neonicotinoids from the effluent of rice paddies (*4*). Neonicotinoid concentrations in Japanese water bodies frequently exceed chronic limits—and sometimes lethal limits—for aquatic invertebrates measured in laboratory studies. The highest concentrations of two neonicotinoids (thiamethoxam and imidacloprid), sampled weekly for 1 year in the Sagami River of the Tokyo metropolitan area, were 0.202 and 0.104 μg L^{-1}, respectively (*4*). Neonicotinoid exposure of 48 aquatic invertebrate species belonging to 12 orders revealed that chronic neonicotinoid concentrations >0.035 μg L^{-1} or acute concentrations >0.200 μg L^{-1} negatively influence highly sensitive aquatic invertebrate species (*5*). The neonicotinoid concentration in the Sagami River exceeds the acute concentration for highly sensitive organisms.

Sensitive organisms include multiple aquatic invertebrates that represent a substantial dietary component of many fish species. Studies using rice paddy mesocosms showed that the pesticide imidacloprid has pronounced effects on aquatic insects (*6*). However, the effects of neonicotinoids on aquatic ecosystems receiving water from rice paddies have not been investigated; this is in part because even taxonomically and functionally similar species differ greatly in their sensitivity to neonicotinoids. Consequently, species within the same genus or family might exhibit a compensatory increase in abundance after the reduction or elimination of a sensitive species. Such functional redundancy, however, is unlikely in ecosystems with few species. Brackish water systems have low species diversity because of the challenges of osmoregulation. Species diversity in oligohaline waters such as Lake Shinji, Japan (fig. S1), our study site, is the lowest among all brackish water systems (*7*).

Benthic invertebrate species in Lake Shinji either disappeared or noticeably declined in abundance between 1982 (before the use of neonicotinoids in the watershed) and 2016, 23 years after continuous application of neonicotinoids began (Table 1). Among the severely affected taxa, the midge *Chironomus plumosus*, an important diet item for smelt (*8*), was absent from all 39 sampling locations in 2016 despite being abundant on the west coast of Lake Shinji in 1982 (fig. S2). *C. plumosus* inhabited Lake Shinji until October 1992, including August, when oxygen depletion often occurs (fig. S3, A and B). However, this species has been collected only infrequently at four long-term sampling points since 1993: in 1998 to 2000 and 2004 to 2005 (fig. S3C and table S1). The possibility that *C. plumosus* abundance was anomalously high in the 1980s and that the drop in 1993 represents a return to baseline conditions is not plausible because *C. plumosus* declined in other lakes at about the same time that neonicotinoids were introduced (e.g., Lake Suwa) (*9*).

In addition to *C. plumosus*, the isopod *Cyathura muromiensis* (fig. S4), the oligohaline polychaete *Notomastus* sp., and oligochaetes (figs. S5 and S6) had all declined in abundance by 2016 (p < 0.05, paired t tests). In contrast, mesohaline polychaetes increased (fig. S7, *Laonome albicingillum*, p < 0.05; paired t test) or remained unchanged (fig. S8, *Prionospio japonica*, p = 0.19; paired t test), probably because their planktonic larva disperse annually to coastal areas after the concentration of neonicotinoids in these areas has declined.

Pelagic invertebrate species in Lake Shinji also declined as neonicotinoids were introduced into its watershed. The biomass of the pelagic copepod *Sinocalanus tenellus*, an abundant zooplankton species in Lake Shinji and an important fish food for smelt, declined after the introduction of imidacloprid to rice paddies in May 1993. The abundance of this species fell from a mean of 108 μg C L^{-1} (n = 137, SD = 119) during the period of May 1981 to April 1993 to 18.2 μg C L^{-1} (n = 143, SD = 22) during the period of May 1993 to April 2005 (Fig. 2; p < 0.0001, unpaired t test).

Reduced abundance of numerous benthic and pelagic invertebrates in Lake Shinji between 1982 and 2016 cannot be explained by oligotrophication or other confounding factors, including particulate chemical oxygen demand, surface water chlorinity (an indicator of saline water intrusion), bottom water dissolved oxygen concentrations, sediment organic matter content, or the invasion of new fish species. Multiple water-quality characteristics were measured monthly in Lake Shinji and showed no clear changes before and after the introduction of imidacloprid in May 1993 (fig. S9). For example, average particulate chemical oxygen demand values in the period May 1984 to April 1993 versus May 1993 to April 2005 were 1.73 mg L^{-1} (n = 103, SD = 1.02) and 1.53 mg L^{-1} (n = 144, SD = 0.61) (p > 0.05, unpaired t test), respectively. Chlorinity was 1.82 g L^{-1} (n = 105, SD = 1.02) and 1.99 g L^{-1} (n = 144, SD = 1.06), respectively, for these two periods (p > 0.05, unpaired t test). There was also no increasing trend in average salinity at the center of the lake between 1982 and 2016 (data not shown). Thus, increased salinity did not cause decreases in annelid density. Dissolved oxygen was 7.45 mg L^{-1} (n = 103, SD = 4.38) and 7.77 mg L^{-1} (n = 140, SD = 3.60), respectively

[1]Institute of Geology and Geoinformation, Geological Survey of Japan (GSJ), AIST, Central 7, Higashi 1-1-1, Tsukuba, Ibaraki 305-8567, Japan. [2]Graduate School of Frontier Science, The University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8563, Japan. [3]Shimane Prefectural Institute of Public Health and Environmental Science, Nishihamasadacho, Matsue 690-0012, Japan. [4]Nagoya City Institute for Environmental Sciences, 5-16-8 Toyota, Minami-ku, Nagoya, Aichi 457-0841, Japan. [5]Creative Engineering Department of Civil and Environment Engineering, Chiba Institute of Technology, 2-17-1 Tsudanuma, Narashino, Chiba 275-0016, Japan.
*Corresponding author. Email: limnologyandoceanography@yahoo.co.jp

($p > 0.05$, unpaired t test), and sediment organic matter content sampled from 15 locations in Lake Shinji did not change significantly between 1982 and 2016 (*10*).

Total neonicotinoid concentration at our sampling site, station ST, near the mouth of a small stream from the rice paddies that are within the watershed of Lake Shinji (fig. S1), was 0.072 μg L^{-1} in June 2018 (fig. S10). This is sufficiently high to induce negative chronic effects on sensitive aquatic invertebrates (*5*). Neonicotinoid concentrations in water samples at station S7, near the mouth of the Hii River, were lower because sampling was done when rainfall inflow was minimal. When neonicotinoid concentrations peaked after rainfall, concentrations at station S7 were potentially as high as those at station ST. Furthermore, at least three types of neonicotinoids (imidacloprid, clothianidin, and thiamethoxam) were detected in the surface water of the west lake basin (station S7) after rice planting in May 2018. Toxicity of multiple neonicotinoids seems to be synergistic in aquatic systems. Using experimental in situ enclosures, chronic toxicity of imidacloprid, clothianidin, and thiamethoxam (and their mixtures) to natural aquatic insect communities showed that the cumulative emergence of insects after the mixture treatments was 42%, 20%, and 44% lower, respectively, than when the three neonicotinoids were applied separately (*11*). No reports are available on the synergistic toxicity of neonicotinoids on *C. plumosus*, but mixtures of neonicotinoids have probably negatively influenced *C. plumosus*, other macrobenthos, and zooplankton such as *S. tenellus*.

Reductions in the abundance of invertebrates, particularly zooplankton, were associated with significantly lower landings of smelt (*Hypomesus nipponensis*) and eel (*Anguilla japonica*) in Lake Shinji after the introduction of neonicotinoid use in 1993 (Figs. 1 and 3). Average annual yields of smelt dropped from 240 tons (n = 12, SD = 57) to 22 tons (n = 12, SD = 53) between the periods 1981 to 1992 and 1993 to 2004 ($p < 0.001$, unpaired t test). For eel, mean annual yield declined from 42 tons (n = 12, SD = 9.7) to 10.8 tons (n = 12, SD = 4.8) between the same periods ($p < 0.001$, unpaired t test). By contrast, ice fish (*Salangichthys microdon*) yield remained unchanged before and after the introduction of neonicotinoids; average annual yield in 1981 to 1992 was 40.9 tons (n = 12, SD = 18) and from 1993 to 2004, it was 53.3 tons (n = 12, SD = 50) (p = 0.43, unpaired t test).

Under the Fishery Act [http://faolex.fao.org/docs/pdf/jap1710JAP.pdf (in Japanese)], the Lake Shinji Fisheries Cooperative Association has the fishing rights for these species and is obliged to increase their abundance. To increase smelt and eel yield, the Lake Shinji Fisheries Cooperative Association releases eyed eggs of smelt and juvenile eels every year. This stocking occurred throughout the 1990s (fig. S11), yet the same fishing gear (net sets) caught 908 tons of smelt in the 1960s and just 27 tons during 1995 to 1996 (*12*).

Young smelt consume crustacean zooplankton, and they feed proportionately more on epiphytic crustaceans and midges as they grow (*8*, *13*). The decrease in zooplankton biomass (Fig. 2) and *C. plumosus* density by 83% and 100%, respectively, with the introduction of neonicotinoids indicates that decreased smelt abundance at Lake Shinji was indirectly driven by the introduction of neonicotinoids. The same is true for eel, which feed on zoobenthos including polychaetes, oligochaetes, and insects (Chironimidae) (*14*). Eel declines were not caused by a reduction in the number of juvenile eels released into Lake Shinji, as this number did not change before and after the introduction of neonicotinoids (fig. S11). Nor was the decrease of eels caused by lower recruitment of postlarval eels (i.e., glass eels) from the ocean. Larval eel recruitment from the ocean to freshwater bodies in Japan remained relatively constant between 1981 and 2004 (Fisheries Agency: www.jfa.maff.go.jp/j/saibai/attach/xls/unagi-8.xlsx); average annual catches of glass eel in Japan decreased by only 32%, from 25.7 tons (n = 12, SD = 7.5) during 1981 to 1992 to 17.6 tons (n = 12, SD = 5.1) during 1993 to 2004 ($p < 0.05$, unpaired t test).

Ice fish landings were unaffected by the food web changes associated with the introduction of neonicotinoids. Small ice fish (<20 mm total length) in Lake Shinji feed more heavily than smelt or eel on primary producers—diatoms occupy 40% of ice fish gut contents and zoo-

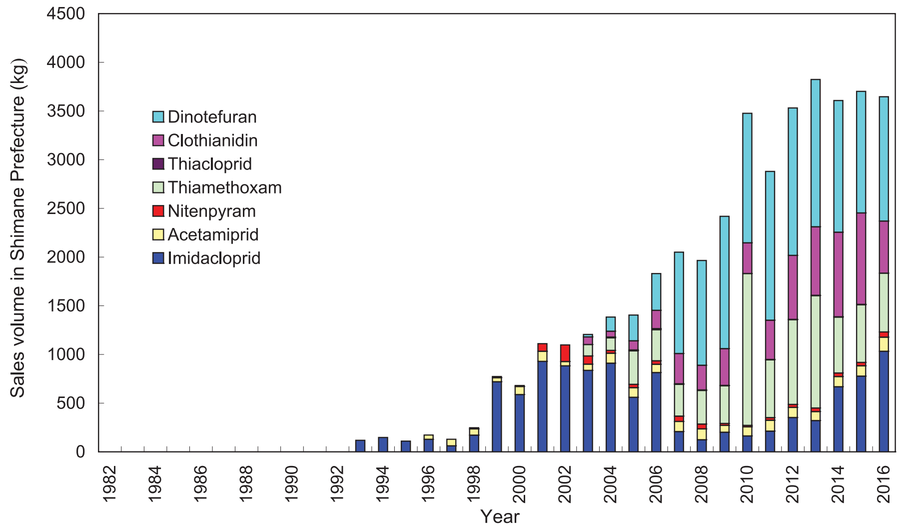

Fig. 1. Quantity of seven kinds of neonicotinoids (kg) sold in Shimane Prefecture, Japan. Data were derived from the "Webkis-Plus: Database of chemicals" (https://www.nies.go.jp/kisplus/).

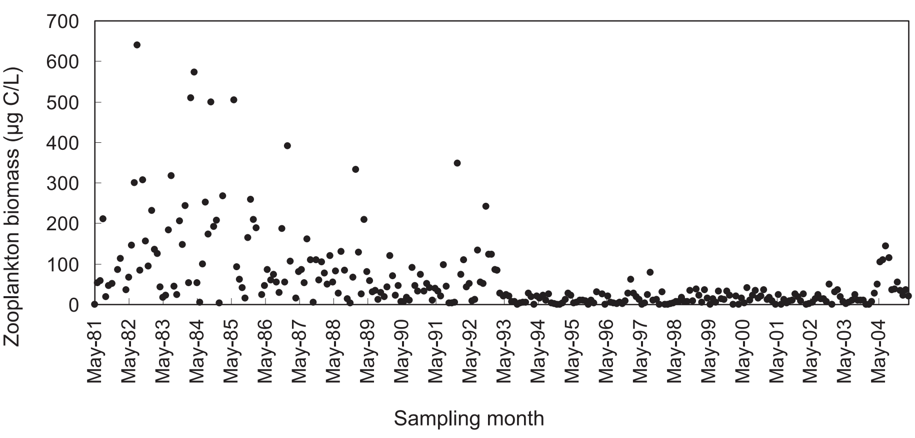

Fig. 2. Zooplankton biomass (μg C L^{-1}) collected monthly at the center of Lake Shinji from May 1981 to April 2005 (data were supplied by the Izumo River Office). Neonicotinoid application began in 1993.

Table 1. Mean (SD) density (individuals m^{-2}) of dominant macrobenthos at 39 locations in Lake Shinji, Japan, sampled in both 1982 and 2016. p values are the result of paired *t* tests. A list of all species collected in 2016 is provided in table S2.

Taxa	1982	2016	p
Arthropods			
Chironomus plumosus	121 (268)	0.0	0.008
Tanypodinae spp.	125 (169)	19 (25)	0.001
Cyathura muromiensis	30 (75)	0.2 (1.1)	0.017
Annelids			
Prionospio japonica	88 (150)	131 (131)	0.192
Notomastus sp.	101 (243)	0.4 (0.8)	0.014
Laonome albicingillum	4.2 (15)	12 (13)	0.026
Oligochaeta spp.	188 (387)	14 (39)	0.007

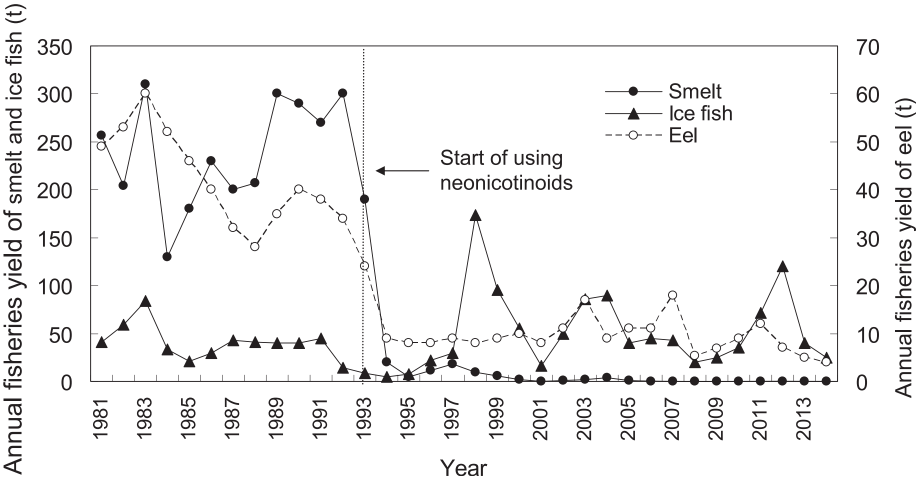

Fig. 3. Annual yield (tons) of smelt, ice fish, and eel in Lake Shinji from 1981 to 2014. The vertical dashed line indicates when neonicotinoid use began in the watershed of the lake.

plankton 60% (*15*). Although larger size classes of ice fish (>21 mm total length) feed mainly on zooplankton, the ability of young ice fish to utilize diatoms may sustain recruitment.

In Lake Shinji, neonicotinoids indirectly reduced fishery yields by decreasing the abundance of invertebrates that serve as food for smelt and eels. Nationwide decreases in fishery yields in the lakes of Japan were also probably caused by food web disruption from neonicotinoids after the widespread use of these pesticides. Neonicotinoids can also affect fish directly. Sublethal effects on fish have been observed from exposure to environmentally relevant concentrations of fipronil, imidacloprid, and thiacloprid (*16*).

Neonicotinoids are the most widely used class of insecticides, representing more than 25% of the global pesticide market, and were valued at more than $3 billion U.S. in 2014 (*17*). Decreased survival, growth, and reproduction of freshwater organisms, particularly aquatic insects and crustaceans, by widespread use of neonicotinoids could alter ecosystem functions related to nutrient transfer from primary producers to secondary consumers, including fish (*16*). Decreased farmland bird populations in the Netherlands was associated with the use of neonicotinoids, which reduced the abundance of insect prey (*18*). In 1962, Rachel Carson wrote in *Silent Spring* (*19*), "These sprays, dusts, and aerosols are now applied almost universally to farms, gardens, forests, and homes—nonselective chemicals that have the power to kill every insect, the 'good' and the 'bad', to still the song of birds and the leaping of fish in the streams…" The ecological and economic impact of neonicotinoids on the inland waters of Japan confirms Carson's prophecy.

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SUPPLEMENTARY MATERIALS

science.sciencemag.org/content/366/6465/620/suppl/DC1
Materials and Methods
Figs. S1 to S11
Tables S1 and S2
References (*20–24*)

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Cascading effects of pesticide use

It is now well known that neonicotinoids negatively affect pollinators. As research has expanded, it has become clear that these globally used insecticides directly affect other ecosystem components, including vertebrates. Yamamuro *et al.* now show that these compounds are indirectly affecting species through trophic cascades (see the Perspective by Jensen). Since the application of neonicotinoids to agricultural fields began in the 1990s, zooplankton biomass has plummeted in a Japanese lake surrounded by these fields. This decline has led to shifts in food web structure and a collapse of two commercially harvested freshwater fish species. The authors argue that such dynamics are likely occurring widely.

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