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**Neonicotinoid Contamination in Tap Water of Akita City, Japan, and
Removal Efficiency of Household Water Purifiers**

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Abstract

Contamination of tap water by neonicotinoid insecticides (NNIs) and the mitigation potential of household purifiers were evaluated in Akita City, Japan. During the peak application period, all NNIs except nitenpyram were detected, with dinotefuran and sulfoxaflor dominating (max. 3063 and 511 ng L⁻¹, respectively), whereas clothianidin, dinotefuran, imidacloprid, and thiamethoxam predominated during the non-application period. The mean Σ NNI concentration in August (2768 ng L⁻¹) exceeded the European Union drinking water standard by more than 20-fold. This study provides the first evidence of sulfoxaflor occurrence in tap water and reports the highest concentrations observed to date, together with a threefold increase in peak dinotefuran levels relative to the previous year, indicating escalating contamination pressure. Household water purifiers with activated carbon exhibited highly variable removal efficiencies. Near-complete or complete NNI removal was achieved by purifiers with activated carbon plus UV or glass cellulose fiber, while other activated carbon units required cartridge replacement and slower flow rates to ensure high performance. In addition, flow-rate reduction is a key consideration for cost-effectiveness. Estimated daily intakes of NNIs from tap water ingestion were highest for infants but remained below health-based guideline values, suggesting no health concerns from NNIs in tap water. Nonetheless, elevated seasonal NNI concentrations in tap water, together with cumulative dietary exposure, potential mixture effects, and unassessed metabolites, highlight the need for enhanced monitoring and updated standards.

Key Words: *activated carbon, cartridge replacement, dinotefuran, flow rate, sulfoxaflor.*

ဂျပန်နိုင်ငံ၊ အာဘီတမြို့ရှိ ပိုက်ခေါင်းရေ၌ Neonicotinoid အင်းဆက်သတ်ဆေးများ ညစ်ညမ်းပါဝင်နေခြင်းနှင့် အိမ်သုံးရေ သန့်စင်စက်များ၏ ဖယ်ရှားနိုင်စွမ်း

သင်းသီရိအောင်၊ လက်ထောက်ညွှန်ကြားရေးမှူး

စာတမ်းအကျဉ်း

ဂျပန်နိုင်ငံ၊ အာဘီတမြို့ရှိ Tap Water တွင် Neonicotinoid အင်းဆက်သတ်ဆေးများ (NNIs)၏ ရာသီအလိုက် ညစ်ညမ်းမှုနှင့် အိမ်သုံးရေသန့်စင်စက်များ၏ ဖယ်ရှားနိုင်စွမ်းကို အကဲဖြတ် လေ့လာခဲ့ ပါသည်။ NNI အသုံးအများဆုံးကာလအတွင်း Nitenpyram မှလွဲ၍ ကျန် NNIs အမျိုးအစားများ အားလုံးကို Tap Water တွင် တွေ့ရှိရပြီး Dinotefuran နှင့် Sulfoxaflor တို့က အဓိက ပါဝင်ကာ အမြင့်ဆုံး ပြင်းအားအနေဖြင့် 3063 ng L^{-1} နှင့် 511 ng L^{-1} အထိ အသီးသီး တွေ့ရှိခဲ့ရပြီး Clothianidin၊ Dinotefuran၊ Imidacloprid နှင့် Thiamethoxam တို့အား NNI အသုံးမပြုသော ကာလတွင် တွေ့ရှိရ ပါသည်။ ဩဂုတ်လအတွင်းရှိ NNI စုစုပေါင်း၏ ပျမ်းမျှ ပြင်းအား (2768 ng L^{-1}) သည် ဥရောပသမဂ္ဂ၏ သောက်သုံးရေ စံချိန်စံညွှန်းထက် အဆ ၂၀ ပိုမို၍ ကျော်လွန်ခဲ့ပါသည်။ ယခုလေ့လာမှုသည် Tap Water တွင် Sulfoxaflor တွေ့ရှိမှုကို ပထမဆုံး အကြိမ် အစီရင်ခံ တင်ပြခြင်းဖြစ်ပြီး ယခင်နှစ်နှင့် နှိုင်းယှဉ်ပါက Dinotefuran ၏ အမြင့်ဆုံး ပြင်းအားမှာ သုံးဆခန့် တိုးလာကြောင်းကို တွေ့ရှိရသဖြင့် ညစ်ညမ်းမှု ပို၍ မြင့်မားလာနေသည် ကို ညွှန်ပြနေပါသည်။ Activated Carbon အသုံးပြုထားသော အိမ်သုံးရေသန့်စင် စက်များ သည် NNI များအား ၆၅ % မှ ၁၀၀ % အထိ ဖယ်ရှားနိုင်ပါသည်။ Activated Carbon နှင့် UV (သို့မဟုတ်) Glass Cellulose Fiber တွဲဖက်သုံးထားသော ရေသန့်စင်စက်များသည် NNI များကို လုံးဝနီးပါး (သို့မဟုတ်) လုံးဝ ဖယ်ရှားနိုင်ပြီး အခြား Activated Carbon စက်များမှာမူ Cartridge လဲလှယ် ခြင်းနှင့် ဖြေးညှင်းစွာရေဖြတ်သန်း စီးဆင်းစေခြင်းတို့ဖြင့် ဖယ်ရှားနိုင်စွမ်းကို သိသိသာသာ တိုးတက်စေနိုင်ပါသည်။ ထို့အပြင်ဖြေးညှင်းစွာရေဖြတ်သန်းစီးဆင်းစေခြင်းသည် ကုန်ကျစရိတ် သက်သာစေနိုင်သော နည်းလမ်းတစ်ခုဖြစ်ပါသည်။ တွက်ချက်မှုများအရ Tap Water မှ တစ်ဆင့် NNIs ကို နေ့စဉ်သောက်သုံးခြင်းကြောင့် ခန့်မှန်း နေ့စဉ် ဝင်ရောက်မှုပမာဏသည် ကျန်းမာရေး စံချိန်စံညွှန်းများ အောက်တွင်သာ ရှိနေသောကြောင့် NNI ပါဝင်သော Tap Water အား သောက်သုံး ခြင်း အားဖြင့် ကျန်းမာရေး ပြဿနာ မဖြစ်ပွားနိုင်ကြောင်း ပြသနေပါသည်။ သို့ရာတွင် Tap Water တွင် NNIs မြင့်မားခြင်း၊ အစားအစာမှ စုစုပေါင်းဝင်ရောက်မှု၊ အခြား ဓာတုပစ္စည်းများမှ ပေါင်းစပ် သက်ရောက်မှုများနှင့် ရေထဲတွင်ပါဝင်နေနိုင်သည့် ဆန်းစစ်ခြင်းမလုပ်ထားသော အသွင်ပြောင်း ပစ္စည်းများကိုထည့်သွင်းစဉ်းစားပါက စောင့်ကြပ်ကြည့်ရှုမှုကိုတိုးမြှင့်ရန်နှင့် သောက်သုံးရေ ဆိုင်ရာ စံချိန်စံညွှန်းများ ပြင်ဆင်ရန် လိုအပ်နေကြောင်းကို ညွှန်ပြနေပါသည်။

Neonicotinoid Contamination in Tap Water of Akita City, Japan, and Removal Efficiency of Household Water Purifiers

1. Introduction

Neonicotinoids (NNIs) are widely used insecticides, particularly in rice paddy agriculture. Their high water-solubility and low octanol–water ($\log K_{ow}$) and soil adsorption ($\log K_{oc}$) coefficients confer high environmental mobility and persistence (Morrissey et al., 2015). Consequently, NNIs are readily transported via surface runoff and groundwater leaching, especially following heavy rainfall and in soils with low organic matter content (Wood & Goulson, 2017). In addition, limited removal efficiencies in conventional wastewater treatment processes for NNIs result in the discharge of NNIs into aquatic environments (Campo et al., 2013; Sadaria et al., 2016). As a result, NNIs are increasingly detected in surface waters and other environmental waters (Berens et al., 2021; Bonmatin et al., 2019, 2021; Casillas et al., 2022; Hano et al., 2019, 2020; Hayashi et al., 2017, 2021; Iancu et al., 2019; Kim et al., 2021; Mahai et al., 2021; Putri et al., 2022, 2023; Sánchez-Bayo & Hyne, 2014; Sato et al., 2016; Struger et al., 2017; Sugino et al., 2023; Thompson et al., 2021; Wan et al., 2019–2021; Yamamuro et al., 2019) and in drinking water sources worldwide (He et al., 2021; Kamata et al., 2020; Kim et al., 2021; Klarich et al., 2017; Luo, 2024; Mahai et al., 2021; Sato et al., 2016; Sultana et al., 2018; Thompson et al., 2021; Wan et al., 2019, 2020 & 2021;). Chronic exposure to NNIs has been associated with neurodevelopmental disorders and other adverse health risks (Carmichael et al., 2014; Hernández et al., 2008; Keil et al., 2014; Marfo et al., 2015; Yang et al., 2014). In Japan, municipal drinking water treatment plants often rely on river water sources and commonly lack advanced treatment processes such as activated carbon adsorption, raising concerns regarding tap water safety. Although NNIs have been reported in Japanese rivers and tap water, sulfoxaflor has not yet been documented, and the effectiveness of household water purifiers for NNI removal remains poorly understood.

2. Objectives

- (a) To analyze NNI levels in tap water in Akita City during the peak application period (August) and the non-application period (October, November, and February),
- (b) To evaluate the removal efficiencies of commercially available household water purifiers, and
- (c) To assess potential human health risks.

3. Materials and Methods

3.1. study area

Akita Prefecture, located in the Tohoku region of Japan (Fig. 1), is one of the country's major rice-producing areas, ranking third nationally in 2023 (458,200 tons) and among the top five users of neonicotinoid insecticides (NNIs) (e-Stat, 2023; Webkis-Plus, 2023). Average NNI shipment volumes reached 17.72 tons kL^{-1} during 2019–2021. Akita City, the prefectural capital, is dominated by rice cultivation, which accounts for ~89% of cropland and is practiced once annually during summer (Luo, 2024).

The city is traversed by the Omono River, which drains a 4,710 km^2 watershed, ~16% of which consists of paddy fields. NNIs applied in rice paddies can readily enter surface waters via runoff and drainage. Municipal water treatment plants abstract source water from the Omono River and employ conventional treatment processes without activated carbon. Previous studies reported dinotefuran concentrations up to 111 $ng L^{-1}$ in the Omono River and detected multiple NNIs in Akita City tap water, with highest concentrations observed during summer (Luo, 2024).

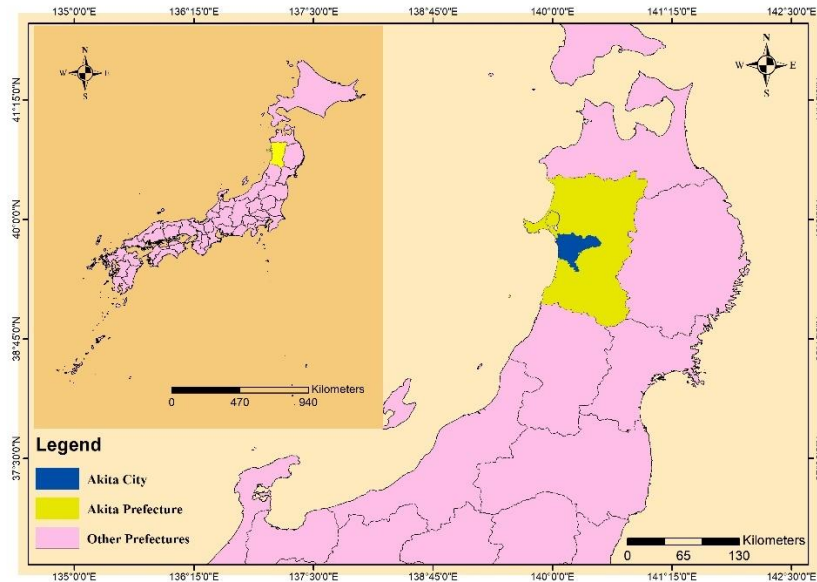


Figure 1. Study area

3.2. sample collection

3.2.1. tap water

Unpurified tap water samples were collected from Akita City using high-density polyethylene bottles during the peak application period (August 2023); Non-application period (October, November 2023, and February 2024). Samples were transported to the Limnology Laboratory, The University of Tokyo (Kashiwa), and stored at 8 °C prior to analysis. Physicochemical parameters during sampling are summarized in Table 1.

Table 1. Information of tap water samples and water parameters

Sampling dates	Water Temp.(°C)	EC (mS/m)	pH	DO (mg/L)	DO Saturation (%)
14 August 2023	/	/	/	/	/
15 August 2023	/	/	/	/	/
16 August 2023	/	/	/	/	/
28 October 2023	16.2	12.4	7.1	10.5	109
31 October 2023	17.2	12.5	6.9	10.1	107
1 November 2023	15.2	13.4	7.3	10.5	109
5 November 2023	17.2	11.8	7.1	10.8	112
20 November 2023	13.8	12.2	7.1	11.0	107
12 February 2024	5.1	13.5	7.2	12.7	102

Note: / means “not measured”.

3.2.2. purified tap water

Purified tap water samples were collected concurrently using household purifiers installed on kitchen taps. Both faucet-mounted and countertop units were evaluated. All purifiers contained activated carbon in combination with other media (e.g., glass cellulose fiber, non-woven fabric, or UV), and differed in cartridge size and usage history (Table 2 and 3).

In August 2023, purified water samples were collected without cartridge replacement or flow-rate adjustment. In October–November 2023, four purifiers were evaluated under four operational treatments: Treatment (1) before cartridge replacement–fast flow, Treatment (2) before replacement–slow flow, Treatment (3) after replacement–fast flow, and Treatment (4) after replacement–slow flow. Fast and slow flow rates ranged from 1.8–4.5 L min⁻¹ and 0.1–1.0 L min⁻¹, respectively (Table 3).

Table 2. Specification of household water purifiers

Purifiers	Types	Materials used in cartridge*	Approximate dimensions of cartridges (cm)		Cartridge Lifespan (Year)	Estimated initial cost** + prices of cartridges (JPY)	Estimated annual cost for cartridges (JPY)
A (Electric)	Countertop	Hollow fibre membrane and anti-bacterial AC	Height	16	2	57,750 + 6,460	3,230
			Diameter	7			
B	Faucet-mounted	AC, non-woven fabric, ceramic (terahertz ore, with plant-derived material-macomo)	Height	10	0.25	20,460 + 5,280	21,120
			Diameter	5			
C	Countertop	AC, glass cellulose fibre	Height	16	1.5	28,800 + 15,800	10,533
			Diameter	12			
D	Faucet-mounted	AC, ceramic and ion exchange fibre, hollow fibre membrane filter	Height	9	0.25	3,780 + 1,496	5,984
			Diameter	5			
E (Electric)	Countertop	Non-woven fabric, compressed AC and UV rays	Height	23	1	86,000+ 35,272	35,272
			Diameter	13			

Notes: *According to the manufacturers' information. ** Price of the whole purifier including a cartridge which can vary depending on the retailer.

Table 3. Information of purified tap water samples and water parameters

Samp ling Dates	Purifiers	% of utilized lifespan*	Flow Rate (L/min)	Water Temperature (°C)	EC (mS/m)	pH	DO (mg/L)	DO Saturation (%)
14-16 Aug 2023	A1 (Electric)	~71	/	/	/	/	/	/
	B1	~50	/	/	/	/	/	/
	B2	/	/	/	/	/	/	/

Samp ling Dates	Purifiers	% of utilized lifespan*	Flow Rate (L/min)	Water Temperatu re (°C)	EC (mS/m)	pH	DO (mg/L)	DO Saturation (%)
	C1	~56	/	/	/	/	/	/
	D	~30 (presuma bly)	/	/	/	/	/	/
28 Oct 2023	A2 (Electric)	~83	F 1.8	16.5	12.4	8.8	10.1	106
			S 0.6	16.6	13.2	9.2	10.2	106
			F 2.3	16.3	13.0	9.0	10.3	107
			S 0.7	16.5	13.7	9.3	10.2	105
1 Nov 2023	C2	~82	F 4.1	15.3	13.4	7.2	10.1	106
			S 0.3	15.8	13.4	7.2	10.2	106
			F 3.7	15.7	13.2	7.5	10.3	107
			S 0.8	15.8	13.3	7.5	10.2	105
5 Nov 2023	E (Electric)	~83	F 3.1	16.8	11.9	7.0	11.5	120
			S 1.0	16.4	11.7	6.9	10.6	107
			F 2.9	16.2	11.7	6.9	10.9	111
			S 1.0	16.6	11.6	6.9	10.4	106
20 Nov 2023	B3	~67	F 4.3	13.8	12.2	7.4	10.9	106
			S 0.1	14.9	12.6	7.6	10.9	109
			F 4.5	13.7	12.6	8.0	11.0	108
			S 0.2	15.0	14.1	8.2	10.7	107

Note: / means “Not Measured”, F means “Fast Flow Rate”, S means “Slow Flow Rate”.

*Utilized lifespan of the purifiers before replacing new cartridges.

3.3. neonicotinoid analysis

Eight NNIs (acetamiprid, clothianidin, dinotefuran, imidacloprid, nitenpyram, sulfoxaflor, thiacloprid, and thiamethoxam) were quantified using solid-phase extraction (SPE) followed by liquid chromatography–tandem mass spectrometry (LC–MS/MS).

200 mL of sample was spiked with isotope-labeled surrogates and extracted using polymeric SPE cartridges. Eluates were concentrated under nitrogen, reconstituted in 20% methanol, and analyzed using a Shimadzu LCMS-8030 triple quadrupole system operated in positive electrospray ionization mode with multiple reaction monitoring (MRM). Chromatographic separation was achieved using a Cadenza CX-C18 UP column (3 µm, 2 mm x 150 mm) and a methanol–ultra pure water gradient containing formic acid and ammonium acetate.

3.4. quality assurance and quality control

All samples were analyzed in duplicate, with coefficients of variation <10%. Procedural blanks, solvent blanks, and spiked blanks were included in each analytical batch, and no target analytes were detected in blanks. Instrument performance and calibration stability were verified periodically using external standards. Method validation, including linearity, detection limits, recoveries, and precision, is described in the Supporting Information.

3.5. removal efficiency of household purifiers

Removal efficiency (%) for each NNI was calculated as:

$$\text{Removal efficiency} = (1 - A/B) \times 100$$

where A and B are NNI concentrations in purified and unpurified tap water, respectively.

3.6. human exposure and risk assessment

Mean concentrations of Σ NNIs were used to calculate the estimated daily intake (EDI) of NNIs through drinking unpurified tap water during the studied months by the following formula (Wan et al., 2019; Thompson et al., 2023):

$$EDI = C \times WIR$$

where EDI is the estimated daily intake of NNIs via unpurified tap water (ng/kg body weight per day), C is the NNI concentration in unpurified tap water from this study (ng/L), and WIR is the water ingestion rates (L/kg body weight per day) focused on different age groups according to USEPA (2011) (Table 4). The EDIs of each neonicotinoid were compared to the WHO and Japan Acceptable Daily Intake (ADI), and chronic reference doses (USEPA cRfD) (Table 8). Age groups were classified into infants (< 1 year old), toddlers (1 to 2 years old), children (3 to 12 years old), adults (>18 years old), seniors (>65 years old).

Table 4. Water ingestion rate

Age Groups	Infants	Toddlers	Children	Teenagers	Adults	Seniors
Water ingestion rate (L/kg. body weight per day)	0.090	0.031	0.029	0.016	0.020	0.021

Source: USEPA (2011) Exposure factors handbook (page: 3-43): Consumer-only estimates of direct and indirect water ingestion.

Researchers suggested using the relative potency factor (RPF) to convert the potency of other NNIs into equivalents of the most predominant NNI (Chang et al., 2018; Wan et al., 2019; Thompson et al., 2023). This allows the EDI determination of each NNI based on their relative potency compared to the most predominant NNI. In this study, dinotefuran was observed as the most prevalent NNI, therefore, RPF of each NNI to dinotefuran was calculated using the following formula (USEPA, 2010):

$$RPF_i = cRfD_{DIN} / cRfD_i$$

where RPF_i is the relative potency factor, cRfD is USEPA chronic reference doses, i is the individual NNI assessed. The calculated RPF values are shown in Table 5.

Table 5. WHO and Japan Food ADI, USEPA cRfD, and calculated Relative Potency Factor (RPF) of NNIs to dinotefuran

	WHO Food ADI (ng/kg. body weight) ^{a,b}	Japan Food ADI (ng/kg. body weight) ^{c,d}	USEPA cRfD (ng/kg. body weight/day) ^e	RPF _i
ACE	70,000	71,000	71,000	0.3
CLO	100,000	97,000	9,800	2.0
DIN	200,000	220,000	20,000	1.0
IMI	60,000	57,000	57,000	0.4
NIT	NA	530,000	NA	-
SUF	50,000	42,000	50,000	0.4
THD	10,000	12,000	4,000	5.0
THM	80,000	18,000	6,000	3.3

Notes: ^a Thompson et al., 2020, ^b FAO/WHO, 2024, ^c Nimako et al., 2023, ^d FSCJ, 2014, ^e USEPA derived cRfD values calculated by Chang et al. (2018) and Thompson et al. (2023).

The concentration of dinotefuran equivalent of NNIs (DIN_{Eq}) was then calculated using RPF_i according to Chang et al. (2018), Wan et al. (2019) and Thompson et al. (2023) as follows:

$$DIN_{Eq} \text{ (ng/L)} = DIN + ACE \times 0.3 + CLO \times 2 + IMI \times 0.4 + SUF \times 0.4 + THD \times 5 + THM \times 3.3$$

DIN_{Eq} was also used to calculate the EDI with average and maximum values of NNIs for the studied months by using the above EDI formula. Risk Quotient (RQ) and Risk Index (RI) of each NNI were calculated according to Thompson et al. (2023) as follows:

$$RQ_i = EDI_i / cRfD_i ; \quad RI = \sum RQ_i$$

where RQ represents risk quotient and RI represents risk index, i represents the individual NNI assessed, EDI represents estimated daily intake of NNIs via unpurified tap water (ng/kg body weight per day), and $cRfD$ is USEPA chronic reference doses. $RI < 1$ indicates no significant health concerns from NNI ingestion through tap water, while $RI \geq 1$ suggests potential health risks.

3.7. statistical analysis

Statistical analyses were performed using R (version 4.3.2). Differences in NNI concentrations among sampling months were assessed using the Kruskal–Wallis test followed by Benjamini–Hochberg–adjusted pairwise comparisons.

4. Results

4.1. seasonal tap water contamination by neonicotinoids

During the peak application period, all NNIs except nitenpyram were detected, with dinotefuran and sulfoxaflor dominating the contamination profile at maximum concentrations of 3063 and 511 ng L⁻¹, respectively while clothianidin, dinotefuran, imidacloprid, and thiamethoxam predominated during the non-application period (Fig. 2). Although the concentrations of all compounds were below the Japan's drinking water standards (See the Supporting Information), the average Σ NNI concentration in August (2768 ng L⁻¹) exceeded the EU Drinking Water Standard (100 ng L⁻¹) by more than 20-fold. To our knowledge, this is the first report of sulfoxaflor detection in tap water. The maximum dinotefuran concentration observed (3063 ng L⁻¹) during the peak application period is the highest ever reported (See the Supporting Information) and represents an approximately threefold increase compared with levels measured in 2022 (868 ng L⁻¹) (Luo, 2024). Significant differences ($p < 0.05$) were observed in Σ NNIs and each compound among the months; except nitenpyram.

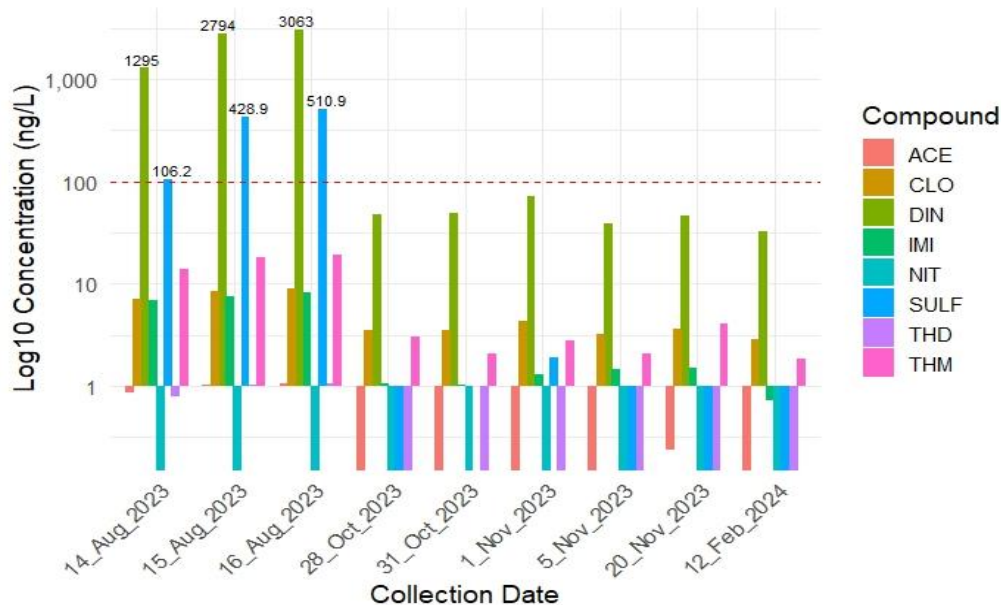


Figure 2. Seasonal Σ NNIs concentrations in tap water, with EU standard (100 ng/L) for comparison.

4.2. household purifier efficiency

Household purifier performance was evaluated under suboptimal operating conditions (no cartridge replacement and unadjusted flow rate) during the peak application period and

under optimized operating conditions (with cartridge replacement and adjusted flow rate) during non-application period.

4.2.1. performance under suboptimal operating conditions

In August 2023, average removal efficiencies for total NNIs differed substantially among purifiers (Fig. 3). Purifier C1 exhibited the highest removal efficiency (99.97%), followed by Purifier D (90.23%), Purifier B2 (85.84%), Purifier B1 (79.95%), and Purifier A1 (69.53%). Differences were observed even between identical products (Purifiers B1 and B2).

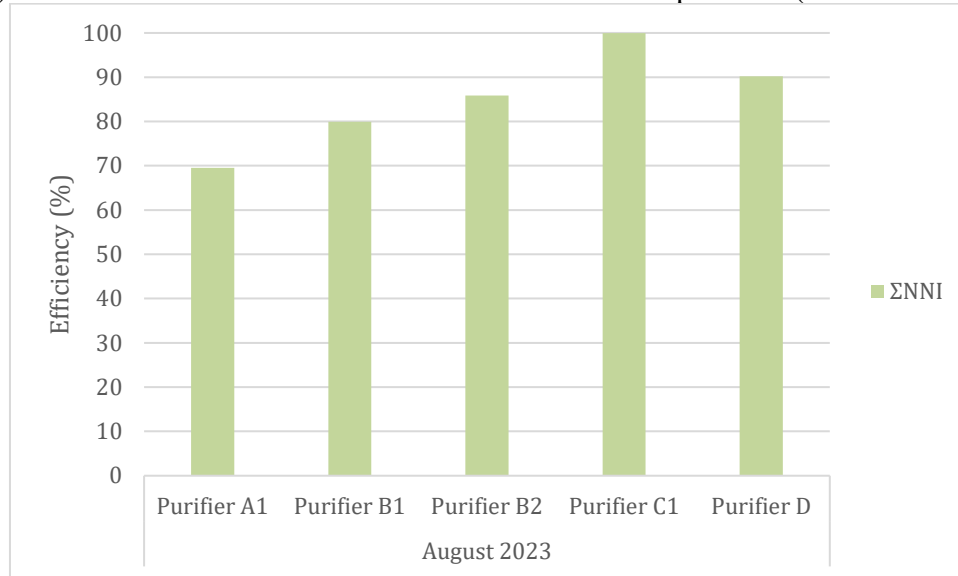


Figure 3. Average efficiencies of household water purifiers from 14 to 16 August 2023

4.2.2. performance under optimized operating conditions

In October and November 2023, purifiers were tested under different operational treatments on separate sampling days (Purifier A2 on 28 October, Purifier B3 on 20 November, Purifier C2 on 1 November, and Purifier E on 5 November 2023). Removal efficiencies varied across purifiers and treatments (Fig. 5).

Purifiers C2 and E consistently achieved high removal efficiencies (99.68% and 100%, respectively) across all treatments. Purifiers A2 and B3 showed moderate removal efficiencies under Treatment 1 (approximately 65% and 70%, respectively), with residual Σ NNI concentrations of $\sim 19 \text{ ng L}^{-1}$ and $\sim 17 \text{ ng L}^{-1}$ (Fig. 4). For Purifier A2, removal efficiency increased to 85% after cartridge replacement and reached 100% under reduced flow conditions. For Purifier B3, removal efficiency increased to 99.24% following cartridge replacement but declined slightly under subsequent flow-rate adjustment treatments.

All tested purifiers effectively removed NNIs when influent concentrations were $\leq 2 \text{ ng L}^{-1}$. At slightly higher concentrations ($\sim 3\text{--}4 \text{ ng L}^{-1}$), removal efficiency depended on cartridge condition and flow rate, particularly for Purifier B3. Considering annual cartridge replacement costs (Table 2) and removal efficiencies under Treatment 2, Purifier A2 followed by Purifiers C2, B3, and E showed the highest cost-effectiveness.

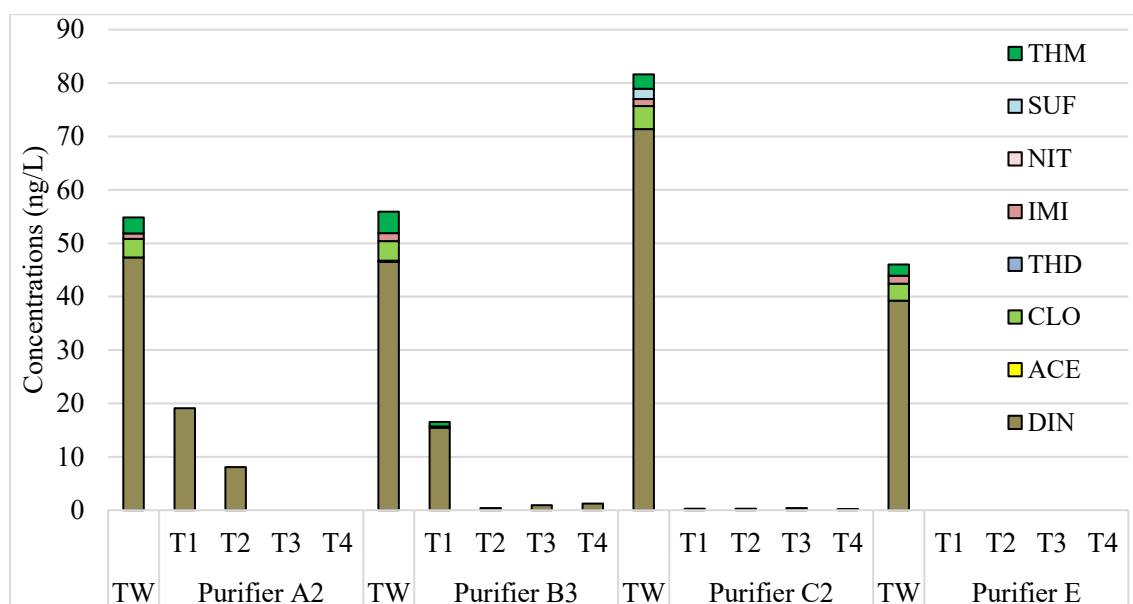


Figure 4. Comparison of NNIs concentrations (ng/L) in unpurified tap water vs purified tap water in different treatments

Notes: TW represents “Tap Water”; T1, T2, T3, and T4 represent “Treatment 1, 2, 3, and 4”.

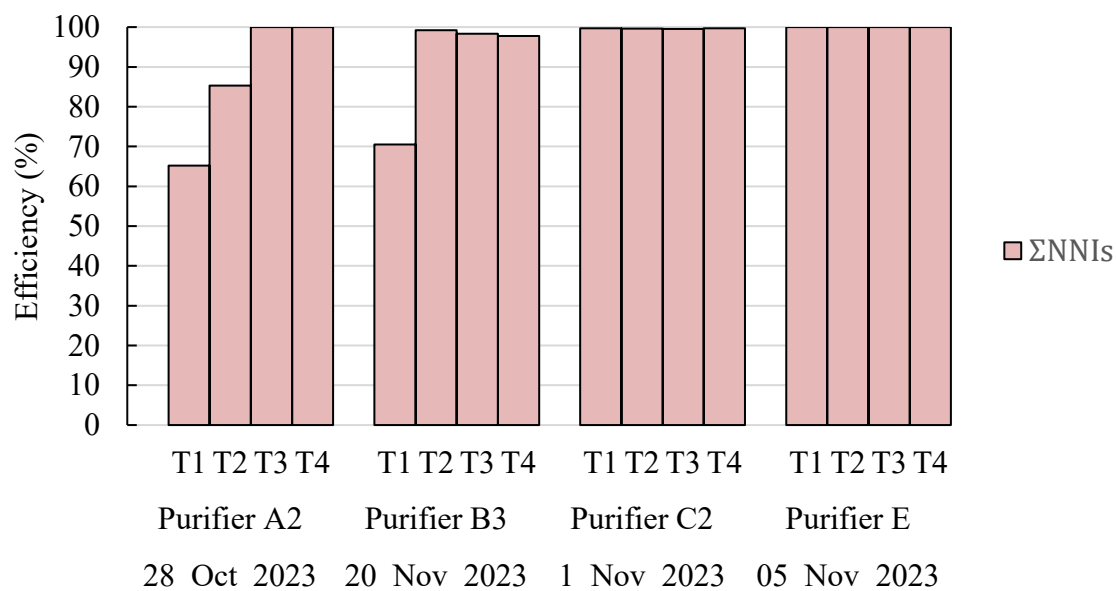


Figure 5. Efficiency of household water purifiers for NNIs removal from tap water

4.3. human exposure and health risk assessment

Infants exhibited the highest estimated daily intake (EDI) of NNIs among all age groups (Table 4). However, all EDIs for Σ NNIs, individual compounds, and dinotefuran equivalents were below the USEPA chronic reference doses, WHO acceptable daily intakes (ADIs), and Japanese food ADIs. Risk quotients for individual NNIs and the overall risk index were consistently below 1, suggesting no health concerns from NNIs in tap water (Table 6 & Supporting Information).

Table 6. Estimated Daily Intake (EDI) (ng/kg.body weight per day) and Risk Index (RI) of NNIs via unpurified tap water ingestion

		Average Σ NNIs					
		Infants	Toddlers	Children	Teenagers	Adults	Seniors
Aug	EDI	249.09	85.80	80.26	44.28	55.35	58.12
	RI	0.012	0.004	0.004	0.002	0.003	0.003
Oct	EDI	5.02	1.73	1.62	0.89	1.11	1.17
	RI	0.00029	0.00010	0.00009	0.00005	0.00006	0.00007
Nov	EDI	5.51	1.90	1.77	0.98	1.22	1.29
	RI	0.00032	0.00011	0.00010	0.00006	0.00007	0.00007
Feb	EDI	3.41	1.17	1.10	0.61	0.76	0.80
	RI	0.00020	0.00007	0.00006	0.00004	0.00004	0.00005

5. Discussion

5.1. factors controlling household purifier performance

The consistently high NNI removal efficiency observed for Purifiers C and E is likely attributable to their cartridge designs, which combine activated carbon with UV treatment in the case of Purifier E (Schreiber et al., 2024), and activated carbon with glass cellulose fiber in the case of Purifier C. Although the independent contribution of glass cellulose fiber to NNI removal has not been clearly demonstrated, combination of cartridge materials could enhance overall removal performance (Schreiber et al., 2024). Collectively, these findings underscore that cartridge composition; rather than activated carbon alone; is a key determinant of effective NNI removal in household water treatment systems.

The lower removal efficiency observed in Purifiers A and B was likely due to extensive cartridge use, consistent with previous findings that prolonged cartridge usage reduces filtration performance. However, reducing the flow rate prior to cartridge replacement substantially improved their removal efficiency.

Differences in efficiency between identical products (Purifiers B1 and B2) further highlight the importance of usage duration and operational conditions.

Overall, these findings demonstrate that household-scale mitigation of NNIs with activated carbon is feasible, though reducing flow rate remain important considerations for cost-effectiveness.

5.2. implications for human health risk

Although all estimated health risks from tap water ingestion alone were below established toxicological thresholds, elevated NNI concentrations in tap water should not be overlooked. Drinking water contributes to cumulative exposure alongside dietary intake from fruits and vegetables, potentially increasing total EDIs beyond those estimated for a single exposure pathway (Chang et al., 2018; Wan et al., 2019).

Moreover, combined exposure to multiple NNIs may result in additive or synergistic effects that are not captured by single-compound risk assessments (WHO, 2017). In addition, drinking water treatment processes may generate metabolites with toxicities comparable to or greater than their parent compounds, which were not explicitly included in this study. Collectively, these findings suggest the need for further evaluation of NNIs in tap water using cumulative and mixture-based risk frameworks.

6. Conclusions

This study demonstrates pronounced seasonal contamination of tap water by neonicotinoid insecticides, including the first detection of sulfoxaflor alongside the highest recorded dinotefuran concentrations, along with a threefold increase in peak dinotefuran levels compared with the previous year. Household water purifiers incorporating activated carbon

with UV treatment or glass cellulose fiber achieved near-complete removal of NNIs, whereas other activated carbon units required cartridge replacement and reduced flow rates for optimal performance. Flow-rate reduction is key to cost-effectiveness. Although estimated daily intakes indicate no immediate health risk from tap water ingestion alone, cumulative exposure from multiple pathways, mixture effects, and potentially toxic transformation products remain concerns. Overall, these findings suggest that existing Japanese drinking water guidelines may not fully capture emerging pesticide risks, underscoring the need for continued monitoring and stricter standards.

This research was presented at the 58th Annual Conference of the Japan Society on Water Environment (JSWE) on March 6, 2024. The disclosure of the findings prompted the citizens' movement to apply activated carbon treatment to the water purification station in Akita City (<https://niida-jousuijo.com/about/>).

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Supporting Information

Table S1. Linearity, MDL and MQL values of target NNIs in tap water

	Calibration equation	R ²	MDL (ng/L)	MQL (ng/L)
ACE	$y = 0.81806 x - 0.00305$	0.999	0.140	0.361
CLO	$y = 0.81600 x - 0.07261$	0.998	0.110	0.284
DIN	$y = 1.35502 x - 0.03227$	0.999	0.141	0.363
IMI	$y = 1.28135 x + 0.04424$	0.999	0.166	0.428
NIT	$y = 0.71191 x + 0.08834$	0.997	0.110	0.284
SUF	$y = 0.9795x - 0.0232$	1.000	0.197	0.507
THD	$y = 0.79356 x - 0.01002$	0.999	0.034	0.088
THM	$y = 0.68884 x + 0.00981$	0.999	0.109	0.281

Table S2. Recovery (%) and precision (%) of the analytical method

Spike Level	0.5 µg/L	10 µg/L	100 µg/L	0.5 µg/L
Guideline	NA	60-115 %	80-110 %	< 30 %
	Recovery (mean ± CV)			Precision
ACE	98 ±3.9	85 ±0.4	99 ±0.7	3.8
CLO	110 ±1.1	88 ±1.5	99 ±1.0	2.5
DIN	93 ±0.6	88 ±0.3	99 ±0.3	1.6
IMI	100 ±2.9	89 ±0.6	96 ± 0.8	4.3
NIT	100 ±1.7	98 ±1.1	88 ±3.1	2.9
SUF	103 ±1.1	70 ±0.9	96 ±2.3	4.7
THD	104 ±0.5	87 ±0.6	99 ± 0.7	0.9
THM	102 ± 1.2	91 ±1.4	96 ±1.0	2.7

Note: NA means “not available”

Table S3. MRM of target NNIs and their surrogates

NNIs and surrogates	Precurs or ion (m/z)	Product ion for quantification (m/z)	Product ion for confirmation 1 (m/z)	Product ion for confirmation 2 (m/z)	Retention time (min)
ACE	223.00	126.00	56.10	99.00	9.99
ACE-d3	226.00	126.00	59.10	99.00	9.97
CLO	249.90	169.10	131.95	113.05	9.34
CLO-d3	253.00	172.05	131.95	113.05	9.33
DIN	203.00	129.10	73.10	114.15	6.00
DIN-d3	206.00	132.10	76.10	116.15	5.97
IMI	256.00	209.00	175.10	128.00	9.20
IMI-d4	259.70	213.05	179.15	214.10	9.16
NIT	271.00	225.10	56.10	126.00	6.38
NIT-d3	273.90	228.10	126.00	257.05	6.38
SUF	278.00	174.05	154.05	105.00	1.00
SUF-d3	285.20	178.05	157.15	109.05	9.98
THD	252.90	126.00	99.00	90.10	11.21
THD-d4	256.90	126.00	99.05	90.10	11.17
THM	291.90	211.05	181.05	132.00	7.83
THM-d3	295.00	214.10	184.10	132.00	7.80

Table S4. Comparison of average (maximum) NNIs concentrations in tap water in this study with those reported in other studies (ng/L).

Location	Samples (n)	Sampling Period	ACE	CLO	DIN	IMI	NIT	SUF	THD	THM	Reference
Akita City, Japan	9	2023 Aug, Oct, Nov and 2024 Feb	0.35 (1.04)	5.06 (8.87)	826.44 (3063.22)	3.30 (8.25)	ND	116.55 (510.93)	0.32 (1.06)	7.51 (19.46)	This study
12 main water supply utilities in Japan	presumably over 7000*	2012 to 2017	/	/ (<10)	/ (2,200)	/ (<100)	/	/	/	/	Kamata et al., 2020
15 drinking water treatment plants in South Korea	45*	June, Aug, and Oct 2020	0.127 (0.696)	ND	23.5 (85.3)	0.580 (1.69)	ND	/	0.008 (0.123)	2.65 (9.07)	Kim et al., 2021
University of Iowa, USA	16	2016 May to July	/	13.99 (33.46)	/	6.81 (26.36)	/	/	/	1.22 (4.15)	Klarich et al., 2017
Iowa City, USA	4	2016 July	/	ND	/	ND	/	/	/	0.39 (0.47)	Klarich et al., 2017
38 Cities of China	84	Between 2017 and 2019	13.20 (69.20)	15.50 (104.00)	32.20 (312.00)	16.20 (68.30)	/	/	12.20 (74.20)	15.60 (214.00)	He et al., 2021
13 sites from Japan	154	May 2022 to April 2023	0.54 (4.42)	3.72 (24.33)	25.28 (868)	1.44 (8.93)	ND	/	0.26 (0.66)	1.85 (15.55)	Luo. 2024

Nakazato Park, Japan	21	April 2014 to Mar 2015	ND	/ (7)	/ (5)	/ (26)	/	/	ND	ND	Sato et al., 2016
32 Provinces of China	789	2019 June	1.75 (182.00)	3.22 (98.80)	0.72 (13.70)	5.63 (233.00)	0.05 (1.62)	/	0.08 (7.18)	6.68 (232.00)	Mahai et al., 2021
6 drinking water treatment plants in Ontario, Canada	12*	2015 July	ND	ND	/	3.97 (4.8)	/	/	ND	43.58 (91.7)	Sultana et al. 2018
Eastern Iowa, USA	47	2018 Dec to 2019 Feb	ND	0.5** (140.5)	ND	<0.09** (29.3)	/	ND	ND	<0.03** (13.1)	Thompson et al., 2023
Wuhan, China	165	2018 Jan, May, July, Oct	5.24 (20.70)	2.03 (8.98)	1.85 (54.80)	12.40 (76.70)	1.07 (14.50)	/	0.05 (0.26)	8.02 (47.00)	Wan et al., 2019
Wuhan, China	159	2019 Feb, April, July, Oct	/	/	/	5.03 (32.0)	/	/	/	/	Wan et al., 2020
Hanoi Vietnam	39	2019 July	/ (0.74)	/ (0.46)	/	/ (2.84)	/	/	/	/ (2.86)	Wan et al., 2021
Four provinces, Vietnam	7	2019 July	/ (0.30)	ND	/	/ (1.45)	/	/	/	/ (1.53)	Wan et al., 2021

Notes: ND: “not detected”, /: “not available or not described in the study”, *: “treated water collected from treatment plants”, **: “median values”.

Table S5. NNIs' regulations for tap water and environmental water

Compounds	NNIs Registration date in Japan	Japan drinking water standard value (ng/L) and (revised year) ^a	Japan environmental water standard value for aquatic plants and animals (ng/L) ^b	EU drinking water regulation ^c (ng/L)
ACE	28 November 1995	200,000 (2013)	2,500	100
CLO	20 December 2001	200,000 (2013)	2,800	100
DIN	24 April 2002	600,000 (2013)	12,000	100
IMI	04 November 1992	100,000 (2013)	1,900	100
NIT	28 November 1995	1,300,000 (2013)	11,000	100
SUF	25 December 2017	NA	30,000	100
THD	26 April 2001	30,000 (2020)	3,600	100
THM	15 August 2000	50,000 (2013)	3,500	100

Notes: ^a[From MHLW \(not available online at present\)](#), ^b MOEJ (2024), ^c Council Directive (2006)

Table S6. Estimated Daily Intake (EDI) of NNIs through unpurified tap water ingestion (ng/kg.body weight per day)

		EDI of Average NNIs					
Date	NNIs	Infants	Toddlers	Children	Teenagers	Adults	
Aug 2023	ACE	0.09	0.03	0.03	0.02	0.02	0.02
	CLO	0.74	0.25	0.24	0.13	0.16	0.17
	DIN	214.56	73.91	69.14	38.14	47.68	50.06
	IMI	0.68	0.23	0.22	0.12	0.15	0.16
	NIT	0.00	0.00	0.00	0.00	0.00	0.00
	SUF	31.38	10.81	10.11	5.58	6.97	7.32
	THD	0.09	0.03	0.03	0.02	0.02	0.02
	THM	1.55	0.54	0.50	0.28	0.35	0.36
	ΣNNIs	249.09	85.80	80.26	44.28	55.35	58.12
Oct 2023	DIN _{Eq}	234.44	80.75	75.54	41.68	52.10	54.70
	ACE	0.00	0.00	0.00	0.00	0.00	0.00
	CLO	0.31	0.11	0.10	0.06	0.07	0.07
	DIN	4.34	1.49	1.40	0.77	0.96	1.01
	IMI	0.09	0.03	0.03	0.02	0.02	0.02
	NIT	0.00	0.00	0.00	0.00	0.00	0.00
	SUF	0.04	0.02	0.01	0.01	0.01	0.01
	THD	0.00	0.00	0.00	0.00	0.00	0.00

Nov 2023	THM	0.23	0.08	0.07	0.04	0.05	0.05
	ΣNNIs	5.02	1.73	1.62	0.89	1.11	1.17
	DIN _{Eq}	5.77	1.99	1.86	1.03	1.28	1.35
	ACE	0.01	0.00	0.00	0.00	0.00	0.00
	CLO	0.34	0.12	0.11	0.06	0.07	0.08
	DIN	4.71	1.62	1.52	0.84	1.05	1.10
	IMI	0.13	0.04	0.04	0.02	0.03	0.03
	NIT	0.00	0.00	0.00	0.00	0.00	0.00
	SUF	0.06	0.02	0.02	0.01	0.01	0.01
	THD	0.00	0.00	0.00	0.00	0.00	0.00
Feb 2024	THM	0.27	0.09	0.09	0.05	0.06	0.06
	ΣNNIs	5.51	1.90	1.77	0.98	1.22	1.29
	DIN _{Eq}	6.34	2.18	2.04	1.13	1.41	1.48
	ACE	0.00	0.00	0.00	0.00	0.00	0.00
	CLO	0.26	0.09	0.08	0.05	0.06	0.06
	DIN	2.92	1.00	0.94	0.52	0.65	0.68
	IMI	0.07	0.02	0.02	0.01	0.01	0.02
	NIT	0.00	0.00	0.00	0.00	0.00	0.00
	SUF	0.00	0.00	0.00	0.00	0.00	0.00
	THD	0.00	0.00	0.00	0.00	0.00	0.00
	THM	0.17	0.06	0.05	0.03	0.04	0.04
	ΣNNIs	3.41	1.17	1.10	0.61	0.76	0.80
	DIN _{Eq}	4.01	1.38	1.29	0.71	0.89	0.94

Table S7. Risk Quotient (RQ) and Risk Index (RI) of NNIs via drinking unpurified tap water

RQ of Average NNIs							
Date	NNIs	Infants	Toddlers	Children	Teenagers	Adults	Seniors
Aug 2023	ACE	1.2x10 ⁻⁶	4.3x10 ⁻⁷	4.0x10 ⁻⁷	2.2x10 ⁻⁷	2.7x10 ⁻⁷	2.9x10 ⁻⁷
	CLO	7.5x10 ⁻⁵	2.6x10 ⁻⁵	2.4x10 ⁻⁵	1.3x10 ⁻⁵	1.7x10 ⁻⁵	1.8x10 ⁻⁵
	DIN	1.1x10 ⁻²	3.7x10 ⁻³	3.5x10 ⁻³	1.9x10 ⁻³	2.4x10 ⁻³	2.5x10 ⁻³
	IMI	1.2x10 ⁻⁵	4.1x10 ⁻⁶	3.8x10 ⁻⁶	2.1x10 ⁻⁶	2.6x10 ⁻⁶	2.8x10 ⁻⁶
	NIT	ND	ND	ND	ND	ND	ND
	SUF	6.3x10 ⁻⁴	2.2x10 ⁻⁴	2.0x10 ⁻⁴	1.1x10 ⁻⁴	1.4x10 ⁻⁴	1.5x10 ⁻⁴
	THD	2.1x10 ⁻⁵	7.4x10 ⁻⁶	6.9x10 ⁻⁶	3.8x10 ⁻⁶	4.8x10 ⁻⁶	5.0x10 ⁻⁶
	THM	2.6x10 ⁻⁴	8.9x10 ⁻⁵	8.3x10 ⁻⁵	4.6x10 ⁻⁵	5.8x10 ⁻⁵	6.0x10 ⁻⁵
	RI	1.2x10 ⁻²	4.0x10 ⁻³	3.8x10 ⁻³	2.1x10 ⁻³	2.6x10 ⁻³	2.7x10 ⁻³
Oct 2023	ACE	ND	ND	ND	ND	ND	ND
	CLO	3.2x10 ⁻⁵	1.1x10 ⁻⁵	1.0x10 ⁻⁵	5.7x10 ⁻⁶	7.1x10 ⁻⁶	7.5x10 ⁻⁶
	DIN	2.2x10 ⁻⁴	7.5x10 ⁻⁵	7.0x10 ⁻⁵	3.9x10 ⁻⁵	4.8x10 ⁻⁵	5.1x10 ⁻⁵
	IMI	1.6x10 ⁻⁶	5.6x10 ⁻⁷	5.3x10 ⁻⁷	2.9x10 ⁻⁷	3.6x10 ⁻⁷	3.8x10 ⁻⁷
	NIT	ND	ND	ND	ND	ND	ND
	SUF	8.9x10 ⁻⁷	3.1x10 ⁻⁷	2.9x10 ⁻⁷	1.6x10 ⁻⁷	2.0x10 ⁻⁷	2.1x10 ⁻⁷
	THD	ND	ND	ND	ND	ND	ND
	THM	3.8x10 ⁻⁵	1.3x10 ⁻⁵	1.2x10 ⁻⁵	6.8x10 ⁻⁶	8.5x10 ⁻⁶	8.9x10 ⁻⁶
	RI	2.9x10 ⁻⁴	1.0x10 ⁻⁴	9.3x10 ⁻⁵	5.1x10 ⁻⁵	6.4x10 ⁻⁵	6.8x10 ⁻⁵
Nov	ACE	1.0x10 ⁻⁷	3.6x10 ⁻⁸	3.3x10 ⁻⁸	1.8x10 ⁻⁸	2.3x10 ⁻⁸	2.4x10 ⁻⁸

2023	CLO	3.4×10^{-5}	1.2×10^{-5}	1.1×10^{-5}	6.1×10^{-6}	7.6×10^{-6}	8.0×10^{-6}
	DIN	2.4×10^{-4}	8.1×10^{-5}	7.6×10^{-5}	4.2×10^{-5}	5.2×10^{-5}	5.5×10^{-5}
	IMI	2.3×10^{-6}	7.8×10^{-7}	7.3×10^{-7}	4.0×10^{-7}	5.0×10^{-7}	5.3×10^{-7}
	NIT	ND	ND	ND	ND	ND	ND
	SUF	1.1×10^{-6}	3.9×10^{-7}	3.7×10^{-7}	2.0×10^{-7}	2.5×10^{-7}	2.7×10^{-7}
	THD	ND	ND	ND	ND	ND	ND
	THM	4.4×10^{-5}	1.5×10^{-5}	1.4×10^{-5}	7.9×10^{-6}	9.8×10^{-6}	1.0×10^{-5}
	RI	3.2×10^{-4}	1.1×10^{-4}	1.0×10^{-4}	5.6×10^{-5}	7.1×10^{-5}	7.4×10^{-5}
Feb 2024	ACE	ND	ND	ND	ND	ND	ND
	CLO	2.6×10^{-5}	9.1×10^{-6}	8.5×10^{-6}	4.7×10^{-6}	5.9×10^{-6}	6.2×10^{-6}
	DIN	1.5×10^{-4}	5.0×10^{-5}	4.7×10^{-5}	2.6×10^{-5}	3.2×10^{-5}	3.4×10^{-5}
	IMI	1.1×10^{-6}	3.9×10^{-7}	3.7×10^{-7}	2.0×10^{-7}	2.5×10^{-7}	2.7×10^{-7}
	NIT	ND	ND	ND	ND	ND	ND
	SUF	ND	ND	ND	ND	ND	ND
	THD	ND	ND	ND	ND	ND	ND
	THM	2.8×10^{-5}	9.6×10^{-6}	8.9×10^{-6}	4.9×10^{-6}	6.2×10^{-6}	6.5×10^{-6}
	RI	2.0×10^{-4}	6.9×10^{-5}	6.5×10^{-5}	3.6×10^{-5}	4.5×10^{-5}	4.7×10^{-5}