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Review

Artificial sweeteners in wastewater treatment plants: A systematic review of global occurrence, distribution, removal, and degradation pathways

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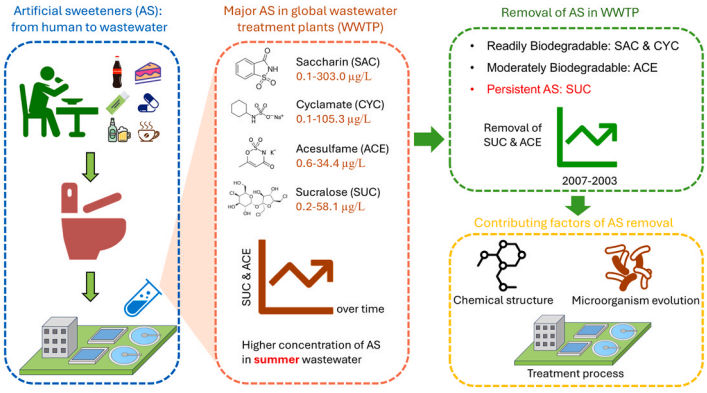
HIGHLIGHTS

- Artificial sweeteners in wastewater at global scale have been assessed.
- Distribution and removal of artificial sweeteners in global WWTPs were evaluated.
- Sucralose in wastewater showed an increasing spatiotemporal trend worldwide.
- Increasing trends of sucralose and acesulfame removal were observed worldwide.
- Future research priorities for artificial sweeteners in wastewater were provided.

GRAPHICAL ABSTRACT



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ABSTRACT

The widespread use of artificial sweeteners in foods, drinks, and pharmaceuticals has led to rising concentrations in wastewater, with specific sweeteners raising concerns due to demonstrated toxicological risks to ecosystems and humans. To date, a comprehensive summary of the occurrence, distribution, and removal status of artificial sweeteners in wastewater treatment plants (WWTP) is lacking, making it difficult to evaluate the associated risks and environmental impacts. We conducted a systematic review of scientific literature and grey literature with rigorous screening covering 24 countries and 6 continents. Globally, sucralose, acesulfame, saccharin, and cyclamate are prevalent artificial sweeteners in WWTP, with concentrations of 0.6–303.0 μg/L in influent and 0.1–81.2 μg/L in effluent. Sucralose showed obvious increasing concentrations over time in wastewater in the United States and Canada, with an increase of 5.6–5.7 μg/L·y in influent and 4.7–5.5 μg/L·y in effluent. Summer wastewater usually contains 11.1–33.3 % higher concentrations of artificial sweeteners than other seasons. Saccharin and cyclamate are the most easily removable sweeteners (>90.0 % removal) in WWTP, followed by acesulfame (25.0–70.1 %) and sucralose (-10.0–10.0 %). Wastewater treatment processes with longer HRT and more diverse microbial communities showed better performance in sucralose removal, while processes with aerobic conditions showed better performance in acesulfame and saccharin removal than anaerobic processes. Increasing trends for persistent sucralose and acesulfame removal have been observed globally, suggesting potential microbial evolution/adaptation. This review contributes to a comprehensive understanding of the

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spatiotemporal distribution and ever-evolving biodegradation of artificial sweeteners in WWTP, providing future perspectives and potential policy requirements.

1. Introduction

Artificial sweeteners are widely used sugar substitutes in processed foods, beverages, drinks, gums, and pharmaceuticals [8,76]. Globally, 8 artificial sweeteners are commonly used, including sucralose, acesulfame potassium (acesulfame-K/acesulfame), aspartame, saccharin, cyclamate, neotame, stevia, and neohesperidin dihydrochalcone (NHDC). These sweeteners share functional groups such as -OH or -NH that interact with human sweet-taste receptors [65], generating a sweetness intensity that is 30–13,000 times that of sucrose (Table 1). Moreover, half of these artificial sweeteners (saccharin, cyclamate, acesulfame-K, and sucralose) are excreted from the human body almost unchanged, with an excretion rate of 70–100 % (Table 1), resulting in negligible/zero calorie burden. Owing to their benefits in the management of weight, diabetes, and tooth cavities [8,76], artificial sweeteners are becoming increasingly popular, with the global market expected to grow from US$7 billion in 2022 to US$12 billion in 2032 [52]. However, the rapid rise in consumption has led to a significant increase in environmental levels of artificial sweeteners [35,37], bringing additional environmental and health concerns [78,88].

Wastewater treatment plant (WWTP) is the primary recipient of artificial sweeteners excreted by humans [78]. Although certain artificial sweeteners (e.g., saccharin and cyclamate) are readily removed in WWTP, majority of artificial sweeteners remain undegraded [47,78], making WWTP a major release source of artificial sweeteners to the wider environment. For instance, sucralose and acesulfame were revealed to be the most predominant micropollutants in effluent wastewater in 90 WWTP across European Union (EU) countries in 2010 [46]. The unremoved artificial sweeteners in effluent flow into surface water, and groundwater, and even end up in drinking water [24,63,86,88], posing toxicity risks to aquatic animals and human health [24,63,86,88]. For instance, sucralose causes embryonic malformation in zebrafish, while high levels of saccharin are neurotoxic to zebrafish [10]. Moreover, the World Health Organization (WHO) has reported that long-term use of artificial sweeteners increases the risk of type-2 diabetes, cardiovascular diseases, and mortality in adults [58]. Therefore, it is essential to comprehensively explore the occurrence, removal, and discharge of artificial sweeteners in WWTP.

To date, a few reviews have focused on artificial sweeteners in surface water, groundwater, and soil/air environments, with regional focus (Europe, Asia, North America) and time-specific analysis [47,48,78]. However, wastewater systems have received limited attention, and the global spatial patterns as well as temporal trends of artificial sweeteners are yet to be systematically explored. A comprehensive summary of the global occurrence, temporal distribution, and removal of artificial sweeteners in WWTP is lacking.

This study systematically summarizes 1) the spatial and temporal distribution of artificial sweeteners in the influent and effluent of WWTP; 2) the removal status of artificial sweeteners in WWTP; 3) the degradation pathway, involved microorganisms, and factors affecting the removal of artificial sweeteners in WWTP. The findings from this review contribute to a comprehensive understanding of artificial sweeteners in WWTP and provide future research priorities.

2. Materials and methods

2.1. Systematic literature review

A systematic search of peer-reviewed journal articles was conducted according to the PRISMA guidelines [50], using three databases (i.e., Web of Science, Scopus, and PubMed), with a detailed search strategy listed in Table S1. Briefly, peer-reviewed articles were searched from two aspects: artificial sweeteners (including synonyms and specific artificial sweeteners) and wastewater. Eight artificial sweeteners were considered in the systematic review. Sucralose, acesulfame-K (hereafter referred to as acesulfame), aspartame, saccharin, and cyclamate were selected for their common use in food, drinks, and PPCPs [33], while stevia, neotame, and NHDC were also included for their emerging use and concerns worldwide [12]. The peer-reviewed article search was conducted in February 2024, with no time restriction.

A comprehensive search of grey literature was also conducted in February 2024, and the search strategy for each database is listed in Table S2, addressing the potential delays between the scientific reporting and observations of artificial sweeteners in WWTP and detection/reporting in authority reports [38]. Seven grey literature databases were searched due to their accessibility and collection of English-language grey literature, including Opengrey.eu, ProQuest Dissertations & Theses Global, the United States of America (USA) Environmental Protection Agency, USA Food and Drug Administration, European Food Safety Authority, WHO Publication, and DART Europe.

The comprehensive inclusion and exclusion criteria for the searched results are shown in Table S3. The search results included published peer-reviewed articles, conference proceedings, and grey literature. Only documents that provided specific concentration of artificial sweetener in domestic wastewater were included. Only the artificial sweetener concentration of wastewater sampled on dry days was included to exclude the potential dilution effect of precipitation. Only documents that provided specific detection methods were included to ensure data accuracy. The concentration of artificial sweeteners was included from multiple wastewater sources, including influent/effluent in domestic WWTP and wastewater in sewer connected to WWTP/wastewater facilities (e.g., wastewater lagoon and pump). The concentration of artificial sweeteners in sewer wastewater was classified as the influent concentration. Only English-language documents were included (both originated and translated).

To ensure search accuracy and minimize bias, three authors participated in the different stages during systematic review, and the details are shown in the supplementary information (SI) Section S1.

2.2. Analytical and visualization methods

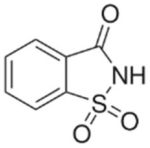
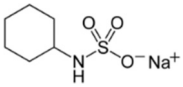
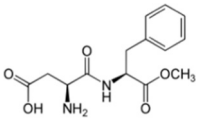
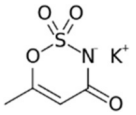
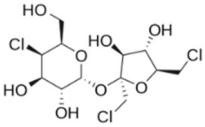
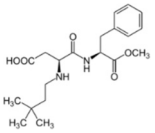
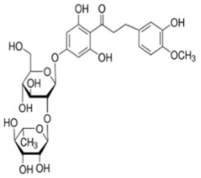
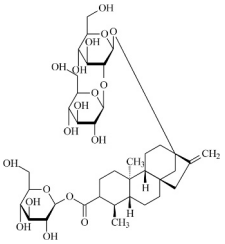
All concentration data of artificial sweeteners were compiled from the included documents using the unit of μg/L. Statistical analyses of the concentration of artificial sweeteners were performed using statistical software R (Version 4.2.2, http://www.R-project.org/). To calculate the seasonal concentration of each sweetener in wastewater, all available data were collected from the included documents in the same countries and seasons. In the Northern Hemisphere, the seasons are defined as spring (March-May), summer (June-August), autumn (September-November), and winter (December-February). In the Southern Hemisphere, the seasons are defined as spring (September-November), summer (December-February), autumn (March-May), and winter (June-August). The annual increase/decrease rate of the concentration of artificial sweetener was calculated using Eq. 1.

$$Annual\ increase \Big/ decrease\ rate = \frac{(\overline{C}_{T1} - \overline{C}_{T0})}{T_1 - T_0} \quad (1)$$

The $\overline{C}_{T_1}$ and $\overline{C}_{T_0}$ represent the average concentration of each artificial sweetener in influent/effluent in the latest year (T_1) and in the earliest year (T_0), respectively.

The removal efficiency of each artificial sweetener was calculated as Eq. 2.

Table 1
The physiochemical characteristics of artificial sweeteners.

Sweetener	Chemical structure	Molecular formula	Log K_{ow}*	Water solubility[a] at 25°C (g/L)	$pK_a^{\#}$	Unchanged compound excretion -rate (%)	Relative sweetness to sucrose	Year of discovery (Country)[d,f]	Aftertaste
Saccharin		$C_7H_5NO_3S$	0.91[a]	0.79	1.3[b]	85–95[d]	500[b]	1879 (the USA)	Bitter and metallic aftertaste[d]
Cyclamate		$C_6H_{12}NNaO_3S$	−1.61[a]	1000	1.7[b]	~70[e]	30–50[e]	1937 (the USA)	Slightly bitter[d]
Aspartame		$C_{14}H_{18}N_2O_5$	0.07[a]	0.564	2.9[b], 7.3	0 [f]	200[b]	1965 (the USA)	Slightly bitter[g]
Acesulfame potassium (Ace-K)		$C_4H_5NO_4S$	−1.33[a]	910	2.0[b]	100[d]	200[b]	1967 (Germany)	Slightly bitter[g]
Sucralose		$C_{12}H_{19}Cl_3O_8$	−1.00[a]	23	−3.0[c], 11.9	100[d]	600[b]	1976 (United Kingdom)	Minimal aftertaste[g]
Neotame		$C_{20}H_{30}N_2O_5$	2.88[a]	0.014	4.1[b] and 7.7	< 2 [f]	7000–13000[b]	1980s (France)	Minimal aftertaste[g]
Neohesperidin Dihydrochalcone (NHDC)		$C_{28}H_{36}O_{15}$	0.75[b]	2	−3.6[c], 9.6	-	1500–1800[f]	1960 (the USA)	Menthol-licorice-like aftertaste[h]
Stevia (Stevioside)		$C_{38}H_{60}O_{18}$	1.20[b]	4.5	−3.6[c], 11.8	0[d]	250–300[b]	1970s (Japan)	Lingering bitter aftertaste[i]

*: Log K_{ow} is the logarithm of the octanol-water partition coefficient. Higher Log K_{ow} means that sweetener tends to adsorb onto organic matter, bioaccumulate in organisms. Lower Log K_{ow} means that tends to stay dissolved in water.
#: pK_a is the negative logarithm of the acid dissociation constant (Ka). Higher pK_a means weaker acid and less ionization at neutral pH. Lower pK_a means that substance is stronger acid and more likely to ionize at neutral pH (~7).
-: Data is not applicable
Source: a:ChemSpider database; http://www.chemspider.com/;
b: PubChem database; https://pubchem.ncbi.nlm.nih.gov/;
c: Human Metabolome database; http://www.hmdb.ca/;
d: Biological fate of low-calorie sweeteners [49];
e: The metabolism of cyclamate to cyclohexylamine in humans during long-term administration [57]
f: Artificial sweeteners–a review [8]
g: Review and synthesis of data on the potential environmental impact of artificial sweeteners [33]
h: Recent analytical methods for the analysis of sweeteners in food: A regulatory perspective [64]
i: Human psychometric and taste receptor responses to steviol glycosides [21]

$$Removal\ efficiency(\%) = \frac{(\overline{C}_{\mathrm{inf}} - \overline{C}_{eff})}{\overline{C}_{\mathrm{inf}}} \times 100 \quad (2)$$

The $\overline{C}_{\mathrm{inf}}$ and $\overline{C}_{eff}$ are the average concentrations of each artificial sweetener in influent and effluent, respectively.

Per capita consumption of artificial sweeteners of included countries was calculated with the details shown in Section S2. All data visualizations were also conducted using R with 'ggplot2' package [44].

3. Results

3.1. Screening and selection of documents

The systematic search yielded a total of 3435 articles from Web of Science, Scopus, and PubMed databases and one document from grey literature (Fig. 1). From the remaining 3099 records, 2930 documents were excluded based on irrelevance, and nine documents were excluded due to inaccessible full text. Then, the remaining 161 documents were further screened through full-text review, and 53 documents were excluded based on the inclusion criteria (Table S3). Ultimately, 108 documents were included for further analysis. Specific artificial sweeteners, their analytical technologies, wastewater sources, served population, wastewater treatment capacity, and countries of included documents are shown in Table S4.

3.2. Global occurrence and distribution of artificial sweeteners in WWTP

Among the included documents, sucralose, acesulfame, saccharin, and cyclamate were the most prevalent artificial sweeteners in influent and effluent wastewater during 2008–2023 (Fig. 2). The occurrence of these four artificial sweeteners in WWTP has been documented in 24 countries across six continents—Asia, Europe, Oceania, Africa, North America, and South America. Among these countries, China (20 documents), the USA (17 documents), Germany (16 documents), Canada (11 documents), and Australia (5 documents) have received the most research attention, accounting for 63.3 % of included documents (Table S4).

The concentrations of artificial sweeteners in influent and effluent have been shown in Fig. 2. Globally, sucralose concentration ranged from 0.6 μg/L to 34.4 μg/L in WWTP influent and from 1.1 μg/L to 27.9 μg/L in WWTP effluent, with the lowest concentrations observed in South Korea (influent) and Vietnam (effluent), and the highest concentrations in the USA (both influent and effluent). Acesulfame concentration ranged from 0.2 μg/L to 58.1 μg/L in influent and from 0.2 μg/L to 35.9 μg/L in effluent, with the lowest concentrations observed in India (both influent and effluent), and the highest concentrations in Spain (influent) and Mozambique (effluent). Saccharin concentration ranged from 5.7 μg/L to 303.0 μg/L in influent and from 0.1 μg/L to 81.2 μg/L in effluent, with the lowest concentrations observed in Vietnam (both influent and effluent) and the highest concentrations in India (both influent and effluent). The notable high

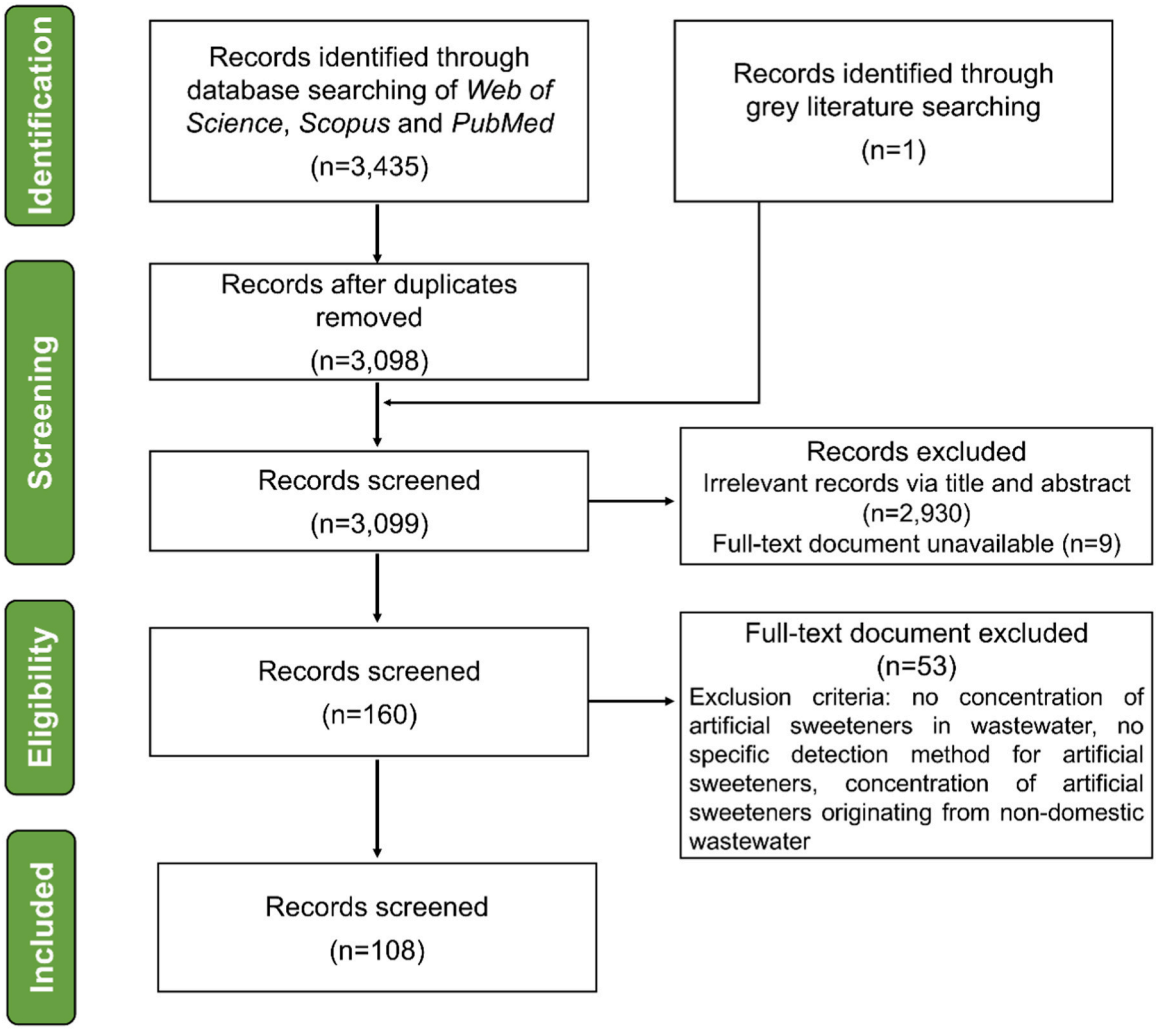


Fig. 1. The process flowchart of systematic review for document screening and selection.

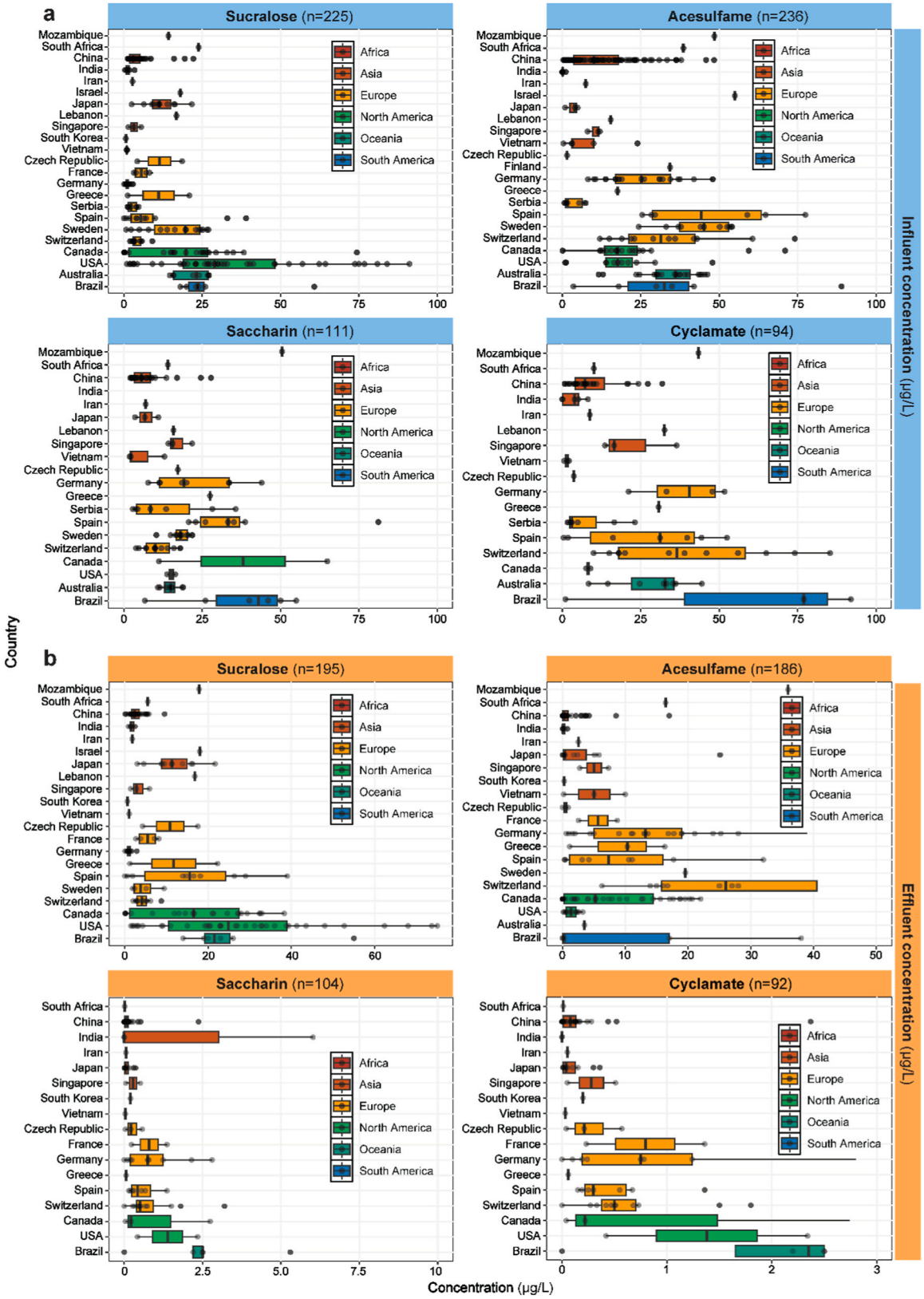


Fig. 2. Artificial sweetener concentrations in influent and effluent wastewater for each country during 2008–2023. (a) influent and (b) effluent. Influent data is only shown in a concentration range of 0–100 μg/L for better visualization. Effluent data is only shown in a centration range of 0–75 μg/L for sucralose, 0–50 μg/L for acesulfame, 0–10 μg/L for saccharin, 0–3 μg/L for cyclamate. The numbers in brackets represent the total number of available data points for that artificial sweetener.

concentration of saccharin in India was likely attributed to the massive consumption of saccharin in India (18.52 mg $d^{-1}p^{-1}$ (milligrams per person per day)) (Fig. S1). Cyclamate concentration ranged from 1.5 μg/L to 105.3 μg/L μg/L in influent and from 0.1 μg/L to 18.2 μg/L in effluent, with the lowest concentrations observed in Vietnam (influent) and South Africa (effluent), and the highest concentrations in Germany (influent) and Mozambique (effluent). Noteworthily, cyclamate was not captured in the USA. This was possibly related to the ban on the use of cyclamate in the USA in the 1970s due to its potential carcinogenic risk [14].

Since aspartame can be metabolized by the human body, it was rarely found in wastewater, and only eight countries detected aspartame in WWTP worldwide (Fig. S2), with an influent range of 0.1–5.3 μg/L and effluent range of 0.1–1.3 μg/L. The highest influent and effluent concentrations of aspartame were observed in Vietnam (5.3 μg/L) and South Korea (1.3 μg/L), respectively. Neotame and NHDC were only captured in WWTP in 3 countries (i.e., China, Singapore, and Serbia) with concentrations below 0.1 μg/L, and no document reported stevia in

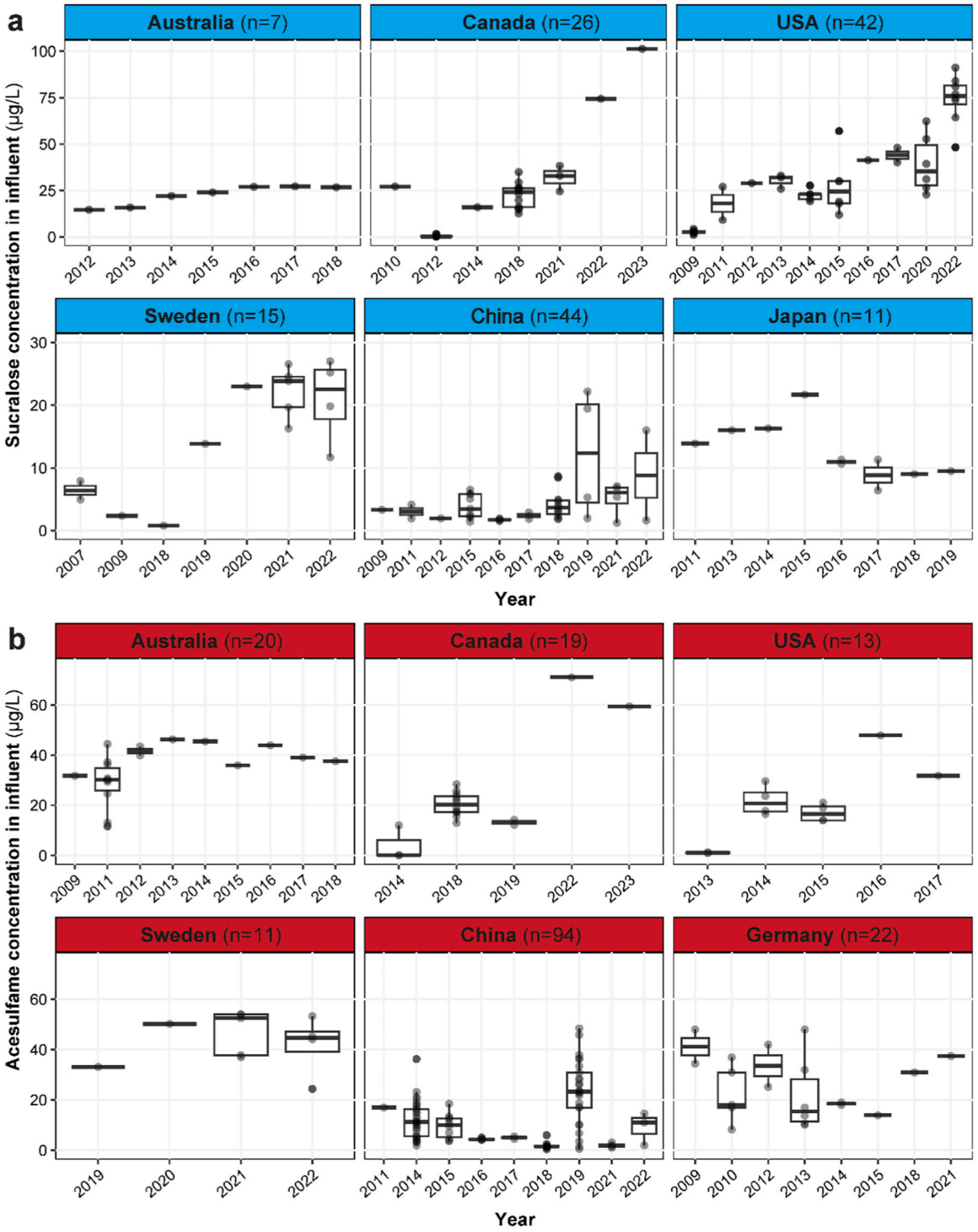


Fig. 3. The yearly influent concentration of artificial sweeteners for countries with available data. (a) sucralose and (b) acesulfame. Single data points at specific years represent the average values reported in the original studies, as the raw data points were not provided in the original studies. The numbers in brackets represent the total number of available data points for that country.

WWTP (Table S4). This was likely associated with their low excretion rates (<2 %) and minimal usage due to high sweetness (Table 1). Also, neotame, stevia, and NHDC were less used than established sweeteners due to the later approval for use in products since the 1990s [84].

Overall, the concentration of artificial sweeteners in influent and effluent in WWTP varied widely depending on the country and the types of artificial sweetener. The differences of artificial sweetener concentration in influent worldwide were likely attributed to the differences in sweetener consumption across countries (Fig. S1), which could be affected by the affordability of artificial sweeteners and preferences of consumers (impacted by factors such as socioeconomic status, sex, and ethnicity) across different countries [60]. The global variance in effluent

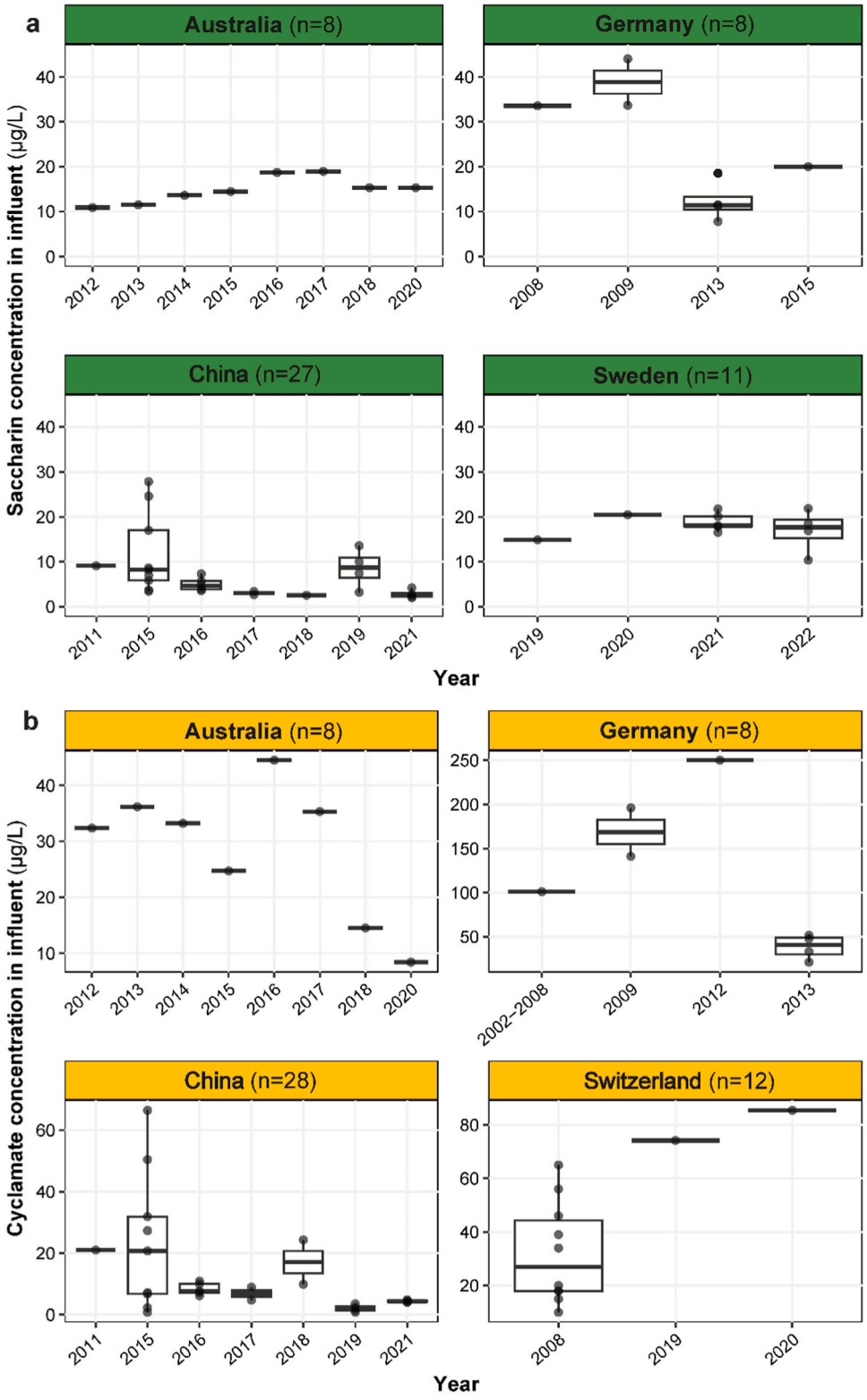


Fig. 4. The yearly influent concentration of artificial sweeteners in Australia, Germany, China, Sweden, and Switzerland (a) saccharin and (b) cyclamate. The numbers in brackets represent the total number of available data points for that country.

concentration of artificial sweeteners was dependent on the differences in the influent concentrations as well as the removal performance of artificial sweeteners in WWTP. The removal ratios of artificial sweeteners were affected by the complexity of sweetener's chemical structure and wastewater treatment processes in WWTP across countries (which will be discussed in the Removal Section 3.5 in detail).

3.3. Temporal distributions of artificial sweeteners in influent wastewater

Temporal variations of artificial sweeteners have also been observed across different countries (Fig. 3). Since the specific year and season information was not provided in all documents, this section only included countries with sufficient data and clear sample collection times, resulting in a total of 501 available data points from eight countries.

Sucralose concentrations in influent increased over time in Australia, Canada, the USA, Sweden, and China (Fig. 3a). The annual increase of sucralose in influent was 2.1 μg/L·y in Australia during 2012–2018, 5.7 μg/L·y in Canada during 2010–2023, 5.6 μg/L·y in the USA during 2009–2022, 1.0 μg/L·y in Sweden during 2007–2022, and 0.4 μg/L·y in China during 2009–2022. The most notable increases were observed in Canada and the USA, while relatively stable sucralose concentration in influent was observed in Japan during 2011–2019 (IQR: 9.3–15.0 μg/L) (Fig. 3a). A significant positive correlation ($R=0.7$, $p = 0.001 < 0.05$) was found between the per capita consumption of sucralose and influent concentration of sucralose, indicating the impact of sucralose consumption on the increasing sucralose concentration in influent. In addition to the increasing per capita consumption, population growth and age group differences in the catchment may also contribute to the higher influent concentration of sucralose, as the total discharge of human excretions containing sucralose increased [36].

Acesulfame concentration in influent increased over time in Australia, Canada, the USA, and Sweden (Fig. 3b). Notable rises of acesulfame in influent were discovered in Canada at 6.6 μg/L·y during 2014–2023, and the USA at 5.6 μg/L·y during 2013–2016, while minor growths of acesulfame concentration in influent were observed in Australia at 0.7 μg/L·y during 2009–2018 and Sweden at 3.85 μg/L·y during 2009–2018. Such increases are consistent with the high consumption of acesulfame in these countries (5.8–13.6 mg $d^{-1}p^{-1}$) (Fig. S1), which is likely due to the high sweetness and slight aftertaste of acesulfame [32,33,59]. Variation in acesulfame concentration in influent was observed in Germany, which decreased from 41.2 μg/L in 2009 to 13.9 μg/L in 2015 and then increased to 37.5 μg/L in 2021 (Fig. 3b). Contrastingly, acesulfame level in influent in China slightly reduced from 17.0 μg/L in 2011 to 9.2 μg/L in 2022 (Fig. 3b), which was likely due to the decreasing use of acesulfame in products [79], and low acesulfame intake in China (3.1 mg $d^{-1}p^{-1}$) (Fig. S1).

Saccharin concentration in influent slightly grew over time in Australia and Sweden (Fig. 4a), with an annual increase of 0.6 μg/L·y in Australia during 2012–2020 and 0.7 μg/L·y in Sweden during 2019–2022, respectively. This might be related to the continuous use and consumption of saccharin in products due to its low price and high sweetness [15,27]. In contrast, saccharin concentration in influent reduced over time in China and Germany, with an annual decrease rate of 0.6 μg/L in China during 2011–2021 and 1.9 μg/L in Germany during 2008–2015. This aligns with their relatively low saccharin consumptions (2.8–6.1 mg $d^{-1}p^{-1}$), which might be due to the unpleasant bitter and metallic aftertaste of saccharin [31].

Downward trends in cyclamate concentration in influent were observed in China and Australia, with an annual decrease rate of 1.7 μg/L·y in China during 2011–2021 and 3.0 μg/L·y in Australia during 2012–2020 (Fig. 4b). Cyclamate concentration in influent in Germany first increased from 101.1 μg/L in 2008 to 250.0 μg/L in 2012 and then decreased to 38.5 μg/L in 2013. The decreasing trends were linked with the lower cyclamate consumption in these countries (Fig. S1), which is likely due to the carcinogenicity and mutagenicity concern of cyclamate on animal in 1969 and the potential influence of the later ban in the USA in 1970 [1,14]. Cyclamate concentrations in influent in Switzerland were higher in 2019 and 2020 compared to 2008 (Fig. 4b). However, due to the limited data points (1 point per year) for 2019 and 2020, this observation may be inconclusive, and further investigation is needed in Switzerland.

In addition, seasonal patterns have been observed in influent concentrations of artificial sweeteners in Australia, China, Germany, and Switzerland (Fig. 5). Summer influent generally showed 11.1–33.3 % higher concentrations of artificial sweeteners than the influent of other seasons, particularly in Australia, Germany, and Sweden (Fig. 5). This was likely associated with the higher consumption of sweetener-added beverages/foods (e.g., iced coke and ice cream) in summer (Fig. S3) [34,91]. In contrast, winter influent in China showed higher concentrations of artificial sweeteners compared to other seasons (Fig. 5). Such seasonal differences might be attributed to long holiday celebrations (e.g., Chinese New Year holiday) in winter (Fig. S3), during which people would consume more sweetened foods/drinks [36].

3.4. Temporal distributions of artificial sweeteners in effluent wastewater

Temporal variations have been observed in effluent concentrations of artificial sweeteners (Fig. S4-S5). Sucralose concentration in effluent showed a similar increasing trend and concentrations as in influent (Fig. S4a and Fig. 3a). The most notable sucralose increases in effluent were observed in Canada and the USA, with an annual increase of 4.7 μg/L·y in Canada during 2010–2023 and 5.5 μg/L·y in the USA during 2009–2022 (Fig. S4a). Relatively stable sucralose concentrations in effluent were observed in China during 2009–2022 (IQR: 1.7–3.4 μg/L) and Japan during 2011–2019 (IQR: 8.9–15.0 μg/L). Such similarity of sucralose concentration and trends in influent/effluent was likely due to the persistence of sucralose in WWTP treatment [78].

Decreasing trends of acesulfame concentration in effluent have been commonly seen in Canada, the USA, China, and Germany (Fig. S4b). The annual decrease rate of acesulfame concentration in effluent was 0.5 μg/L·y in the USA during 2013–2016, 1.1 μg/L·y in China during 2011–2022, 1.4 μg/L·y in Germany during 2009–2021, and 1.2 μg/L·y in Canada during 2008–2023. Since acesulfame concentrations in influent samples increased over time in these countries (Fig. 3), reduced acesulfame concentration in effluent is likely attributed to the increased acesulfame removal performance in wastewater treatment processes, which will be discussed in detail in Section 3.5. Concentrations of saccharin and cyclamate in effluent were observed mostly below 1.0 μg/L without temporal variations (Fig. S5). This is due to the superior degradability of saccharin and cyclamate in WWTP (Fig. 6). Seasonal changes of artificial sweeteners in effluent were not analyzed due to insufficient data.

3.5. Removal of artificial sweeteners in WWTP

The removal performance of artificial sweeteners varied depending on the properties of the artificial sweetener itself, and wastewater treatment processes (Fig. 6). Saccharin and cyclamate were identified as the most easily degradable artificial sweeteners in WWTP, with a high removal rate of 94.3–99.9 % (Fig. 6a). Acesulfame was a moderately degradable artificial sweetener, with a removal rate of −97.8–99.9 % (IQR: 8.8–91.9 %). In contrast, sucralose was the least removed artificial sweetener, with a removal rate of −120.9–99.3 % (IQR: −17.6–20.7 %). The negative removal rate of sucralose observed was mainly attributed to the complex chemical structure and back-release from sludge to wastewater [66,70], while some extreme negative removal rates may arise from the variations in sampling methodology, analytical uncertainties, or WWTP operational fluctuations [55,67].

Among the processes used in WWTP, activated sludge-based (AS) processes, including conventional activated sludge process and modified processes using activated sludge, were predominantly used in 128

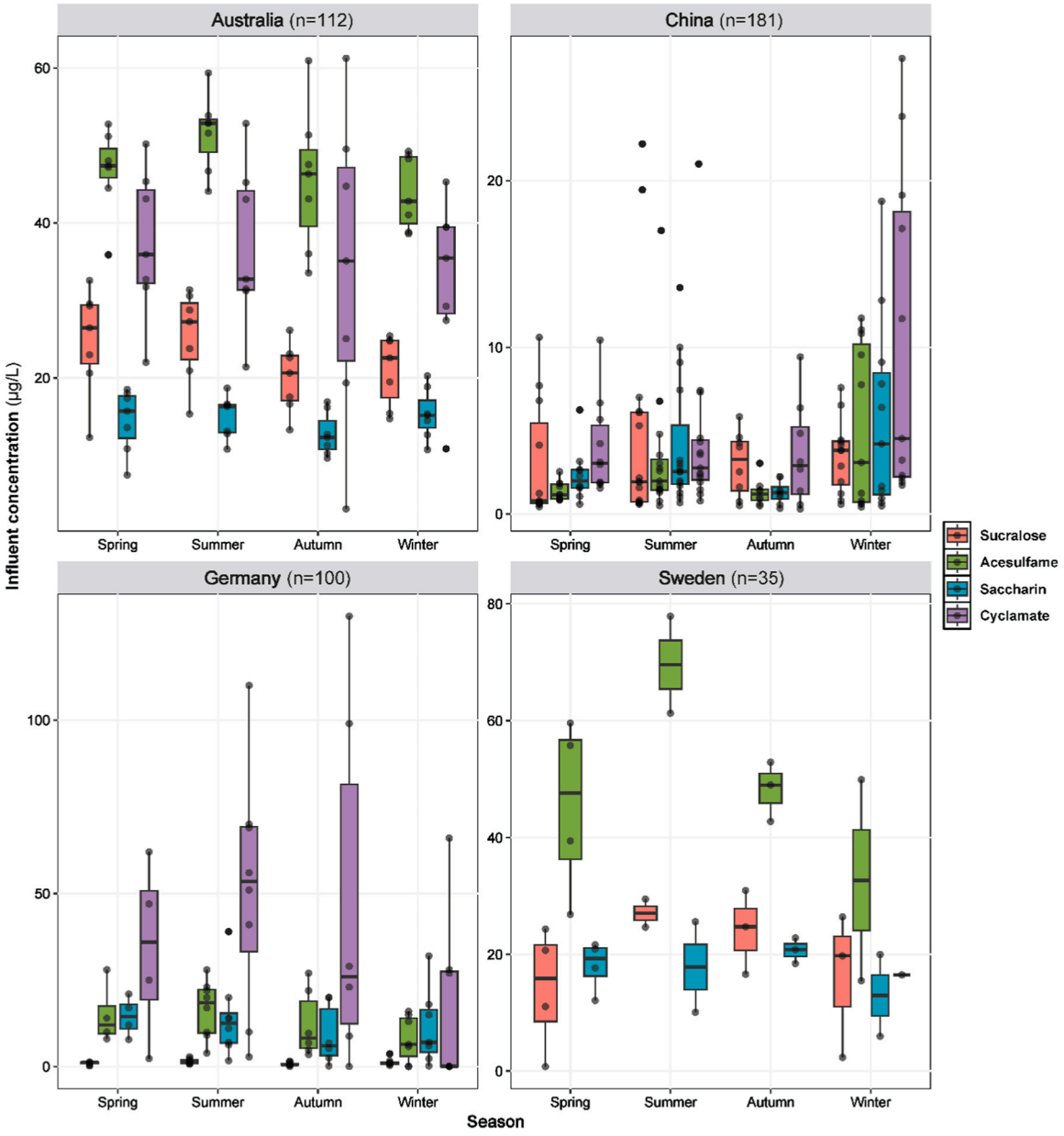


Fig. 5. Seasonal concentrations of artificial sweeteners (μg/L) in influent in Australia, China, Germany, and Sweden. The numbers in brackets represent the total number of available data points for that country.

WWTP (93.4 %), followed by wastewater lagoons in 7 WWTP (5 %), and chemical flocculant-ultraviolet disinfection (UV) approach in 3 WWTP (2 %) (Fig. 6a). Overall, anaerobic-anoxic-oxic (A/A/O) process appeared to be the optimal treatment process for the removal of artificial sweeteners due to its extensive use worldwide (31.3 % of WWTP) and substantial removal capability for most artificial sweeteners (removal IQR: 44.5–94.5 % for acesulfame, 95.6–100 % for saccharin, and 97.6–100 % for cyclamate). All activated sludge-based processes showed superior performance in the removal of saccharin and cyclamate (>90 %) except upflow anaerobic sludge blanket reactor (UASB) in saccharin (38.94 %, IQR: 13.22–64.66 %). Contrastingly, obvious differences have been observed in the removal of persistent artificial sweeteners (i.e., sucralose and acesulfame). The detailed removal rate in each country and the impact of treatment processes on specific sweetener removal will be discussed in the following sections.

3.5.1. *Removal of sucralose*

Sucralose is a refractory and persistent pollutant in WWTP. Globally, 44.4 % of included countries showed negative removal ratios (<0 %) of sucralose in WWTP (Fig. 6b). Such negative removal was primarily due to the stable chlorine atoms in sucralose structure (Table 1), and the release of sucralose from solid phase (i.e., sludge) to wastewater [66, 70]. The lowest removal performance was observed in India [4,5,54], reaching −30.2 % (mean, IQR: −45.4–-11.1 %) (Fig. 6b). This was likely attributed to the low influent level of sucralose concentrations (0.38–3.42 μg/L) and back-release of sucralose in sludge (sucralose concentration in sludge: 0.6–1.9 μg/g) in WWTP in India [70]. Since sucralose is persistent and hardly degraded in WWTP, even a small amount of sucralose back-release from sludge to wastewater during treatment could result in an obvious increase in effluent concentration and high negative removal rates when the sucralose concentration in the influent is low. Comparable removal ratios of sucralose (-10.0–10.0 %) were observed in most countries, except Iran, South Africa, and China

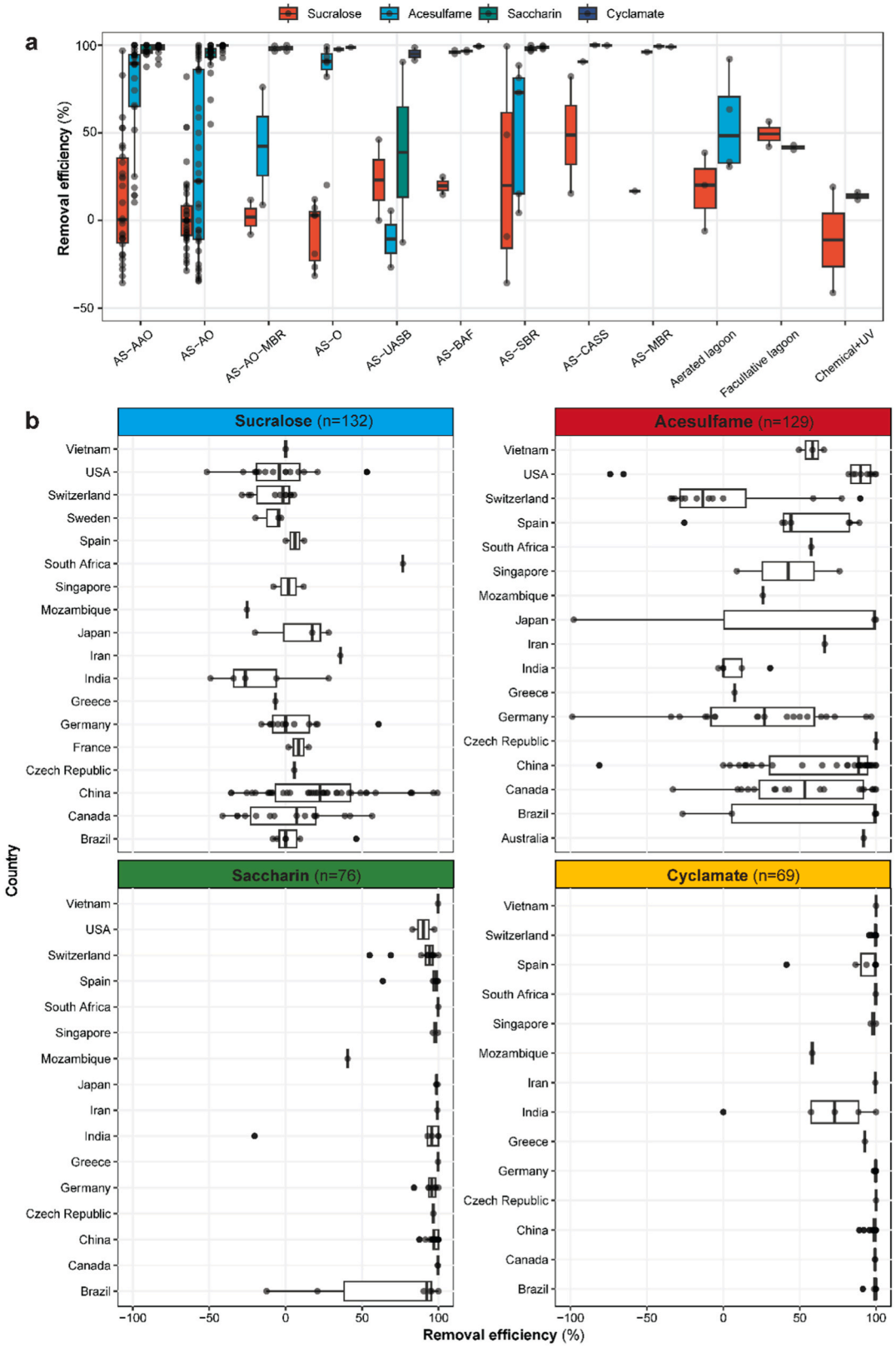


Fig. 6. Removal status of artificial sweeteners. (a) removal efficiency in different wastewater treatment processes, (b) average removal efficiency of artificial sweeteners in included countries (white square means no available data). Activated sludge (AS) processes: anaerobic-anoxic-oxic (A/A/O) process, anaerobic-oxic (A/O) process, oxygen ditch (O), membrane bioreactor (MBR), upflow anaerobic sludge blanket reactor (UASB), biological aerated filter (BAF), sequencing batch reactor (SBR), cyclic activated sludge system (CASS); ultraviolet (UV). The numbers in brackets represent the total number of available data points for that artificial sweetener.

(Fig. 6b). Moderate removal ratio (35.8 %) of sucralose was observed in Iran, likely due to its low influent concentration of sucralose (2.82 μg/L) [72]. One document reported a 76.6 % removal rate in South Africa; however, this observation is inconclusive due to limited data points and insufficient treatment process information, requiring further investigation. China showed a rather notable sucralose removal. Removal ratio of 21.1 % (IQR: −8.7 %-42.2 %) was observed in 38 WWTP, and 4 WWTP showed superior removal ratio of sucralose (>80 %) (Fig. 6b). This is likely due to the diverse microorganisms in the WWTP [19].

Globally, sucralose removal increased from −3.0 % (IQR: −20–9.9 %) before 2017, to 31.3 % (IQR: 6.0–56.7 %) in 2022 (Fig. 7). Such increasing trend is more obvious in China, where the sucralose removal ratios in WWTP increased from −25.4 % (IQR: −35.4 –-19.8 %) in 2016 to 39.0 % (IQR: 24.8–57.4 %) since 2018 (Fig. S6). Such an increase was also observed in other countries, despite lower removal rates. For instance, in Canada, the removal ratio of sucralose in WWTP increased from 2.9 % in 2018 to 15 % in 2023 [56,77], and relatively high removal ratios (38.7–56.5 %) were observed in 3 WWTP [77]. Since these results were all observed in WWTP using biological treatment processes as primary treatment, these increases in sucralose removal suggested the emergence of sucralose biodegradability in WWTP, which was likely due to potential microbial evolution for sucralose removal in WWTP [23]. The treatment process also showed a clear impact on the removal ratios of sucralose. Facultative lagoon treatment showed the highest removal for sucralose (49.3 %, IQR: 45.7–52.9 %), followed by cyclic activated sludge system (CASS) (48.8 %), sequencing batch reactor (SBR) (20.0 %, IQR: −15.8–61.5 %), and aerated lagoon (20.0 %, IQR: 7.0–29.4 %) (Fig. 6a). The high

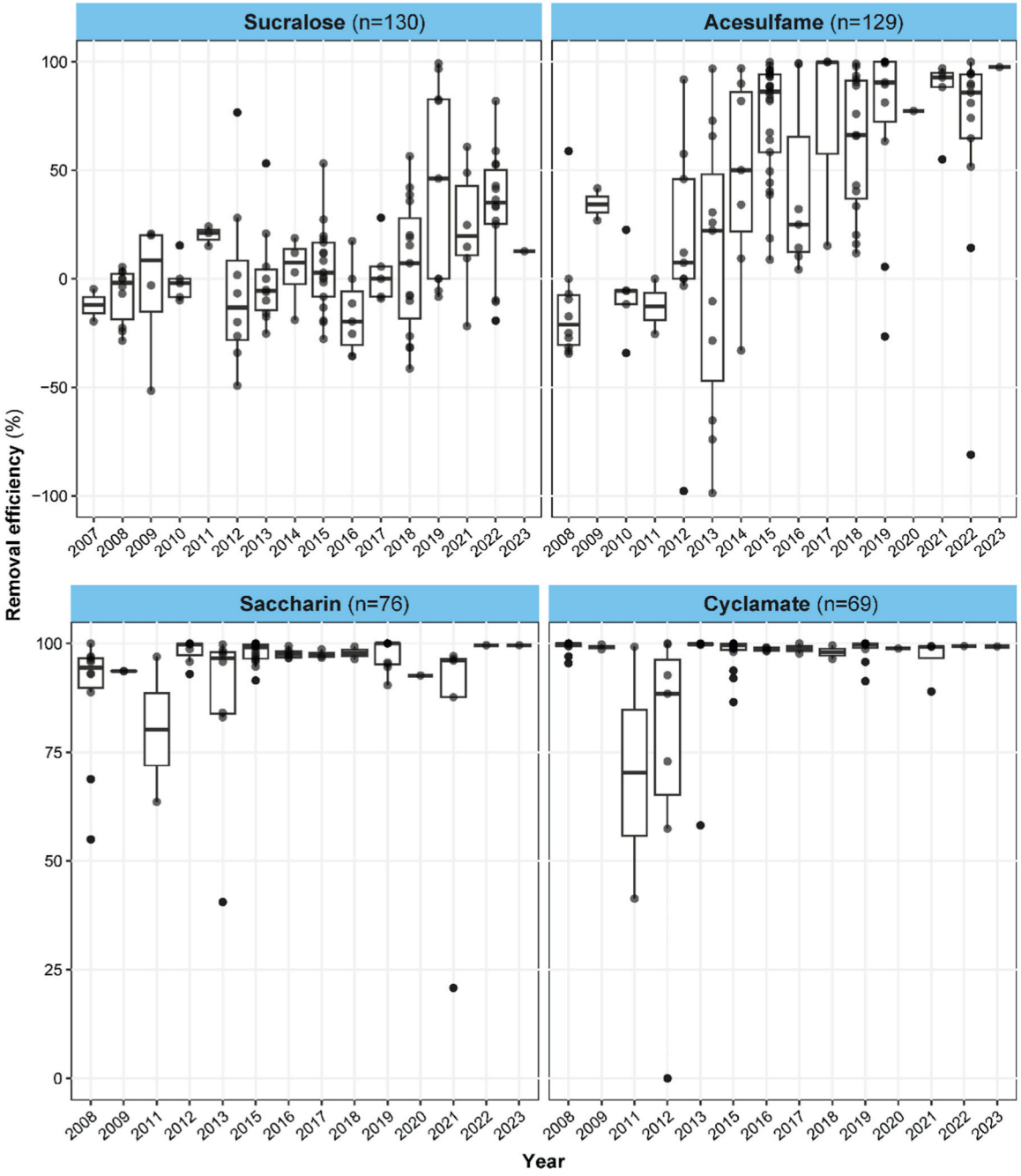


Fig. 7. The yearly removal efficiency of each artificial sweetener in WWTP. The numbers in brackets represent the total number of available data points for that artificial sweetener.

removal ratio in wastewater lagoon was due to its prolonged biodegradation under long hydraulic retention time (20–150 d) [77], while the sucralose removal in CASS and SBR was likely due to the synergistic degradation of aerobic, facultative and anaerobic microorganisms within the same reactor [19]. Additionally, a few recent studies (N = 3) reported higher removal ratios for sucralose [16,19,66]. Removal ratio of 52.7–96.8 % was observed in China through conventional A/A/O process [19,66], and 60.8 % in Germany through anaerobic-oxic (A/O) process [16]. These high removal rates in conventional biological processes indicated the potential microbial evolution for sucralose removal in WWTP.

To date, no study has identified the specific microbial phylotypes or biotransformation products involved in sucralose removal in real wastewater treatment in WWTP. Recent studies have reported the sucralose removal in lab experiments with the sludge from WWTP [23,51]. Using enriched consortia seeded with activated sludge, sucralose was degraded into TP-373N ($C_{12}H_{16}CI_2O_9$) via dechlorination and hydroxylation, TP-409N/TP-433P via oxidation and hydroxylation, and TP-455N via hydroxylation, in a batch bioreactors using sucralose as only carbon source (Fig. 8a) [23]. Similarly, using ethanol as co-substrate, sucralose was degraded by 25.7–52.6 % in anaerobic reactors, which were inoculated with sludge from a Spanish WWTP and a Brazilian WWTP [51]. Genera like *Clostridium* and *Syntrophobacter* were speculated as the contributing phylotypes in sucralose removal [51]. Also, phylotype *Polyangiaceae* and species UBA11579 were suspected of participating in sucralose biodegradation [23]. These results shed light on the potential biotransformation pathway and biodegradation of sucralose in WWTP, while further studies and sequencing evidence in real wastewater treatment plant are still required.

3.5.2. Removal of acesulfame

Most countries (94.1 %) showed positive acesulfame removal worldwide, with mean removal efficiency ranging from 3.4 % to 99.0 % (Fig. 6b). Moderate removal ratios (25.0–70.1 %) of acesulfame were observed in 47 % of countries, while removal rates exceeded 80.0 % in the USA, Czech Republic, China, and Australia (Fig. 6b). A weak positive but not significant correlation (R=0.16, $p > 0.05$) was observed between the influent concentration and the removal efficiency of acesulfame in WWTP. The highest removal (99 %) was observed in a WWTP using biological process with sludge in Czech Republic [30]. Removal rates of 82.0–95.9 % were observed in 9 WWTP in the USA [17,26,67], 74.2–98.5 % in 20 WWTP in China [25,68,87,90,91], and 91.9 % in a WWTP in Australia [69] (Fig. 6b). Although acesulfame was commonly considered as recalcitrant to biodegradation before, recent studies have revealed the microbial adaptation to acesulfame degradation during denitrification in WWTP [28,42,43,7], which were further clarified with

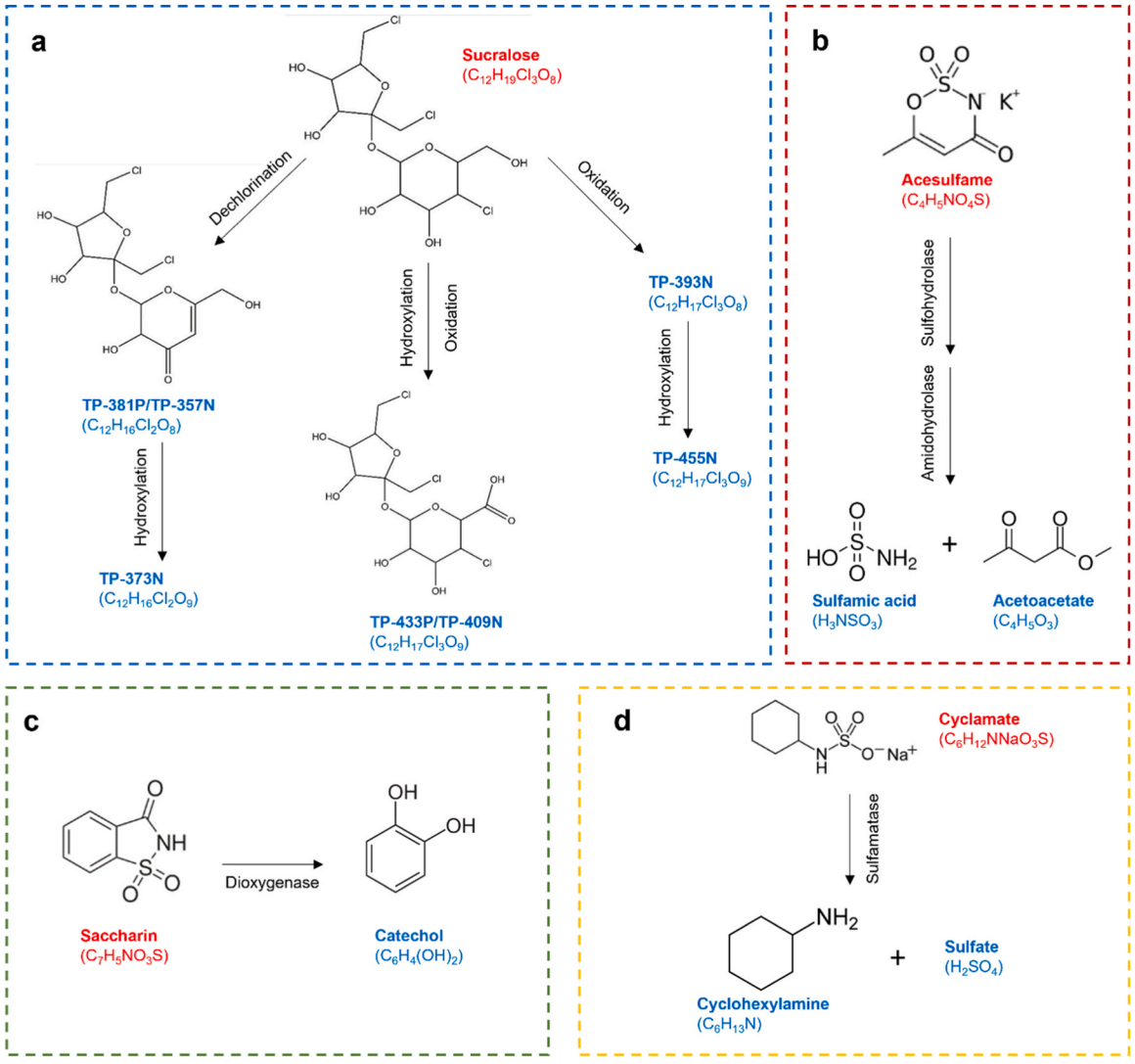


Fig. 8. The potential biotransformation product and process of artificial sweeteners during biodegradation. (a) sucralose (b) acesulfame (c) saccharin (d) cyclamate. Red chemical names are the artificial sweeteners, and the blue chemical names are the potential biotransformation products of artificial sweeteners.

the temporal variation of acesulfame removal and specific microbes later. The lowest acesulfame removal was observed in 6 WWTP in Sweden [3,70], with mean removal efficiency of −65.6 % (Fig. 6b). This was likely due to the release of acesulfame from sludge and the conversion of intermediates alkyl sulfamate (e.g., TP180a and TP180b) [28,7].

Wastewater treatment processes also showed clear impact on acesulfame removal. Biological aerated filter (BAF) showed the highest removal rate for acesulfame (96.0 %) in 2 WWTP, followed by membrane bioreactor (MBR) (95.0 %) in 1 WWTP, oxygen ditch (90.3 %, IQR: 82.0–97.0 %) in 5 WWTP, and A/A/O (89.1 %, IQR: 40.0–94.5 %) in 28 WWTP (Fig. 6a). The superior performances of BAF and MBR might be attributed to the effective biodegradation [87,90], while these observations were only captured in limited WWTP and required more investigation. Since oxygen ditch and A/A/O are basic conventional activated-sludge processes, the high removal rates in oxygen ditch and A/A/O were likely due to the microbial adaptation for acesulfame removal. Also, treatment processes with aeration showed significantly 87.67 % higher removal rates of acesulfame ($p < 0.05$) than USAB (-10.6 %, IQR: −18.6–2.5 %), indicating the necessity of aerobic process in acesulfame biodegradation. Interestingly, the A/A/O process, which incorporates anaerobic, anoxic, and aerobic stages, achieved a significant 66.89 % higher acesulfame removal ($p < 0.05$) than the A/O process, which lacks a dedicated anoxic phase. Since both processes include anaerobic and aerobic stages, while only AAO provides a dedicated anoxic environment for denitrification, this indicated the denitrification step as a major contributor to acesulfame removal.

Moreover, acesulfame removal showed an obvious increasing trend worldwide over the past 15 years (Fig. 7). Globally, acesulfame removal rate was −11.8 % (IQR: −27.3 −-5.4 %) during 2008–2011, then it reached 3.7 % (IQR: −2.4–37.5 %) in 2012, and dramatically surged to 93.5 % (IQR: 83.4–95.5 %) in 2021 (Fig. 7). The initially observed positive acesulfame removals were observed in a Swiss WWTP in early 2008 at 58.8 % [6], followed by a German WWTP in 2009 at 41.7 % [61], both using A/O process. These observations align with the conclusions in Kahl's study that the biodegradation of acesulfame in WWTP emerged around 2010 and then became widespread globally [28]. Therefore, such increases in acesulfame removal were attributed to the microbial evolution/adaptation for acesulfame biodegradation in WWTP. Phylotypes *SM1A02*, *Phyllobacteriaceae*, *Methylophilaceae*, *Bradyrhizobiaceae,* and *Thauera* have been observed as acesulfame degradation-related microorganisms during denitrification [11,28,42]. Extensive studies have revealed that acesulfame was ultimately degraded into inorganic compound sulfamic acid through denitrification/anoxic microbial process in WWTP [28,42,43,7], and this biotransformation was achieved through sequential action of sulfohydrolase and amidohydrolase enzyme (Fig. 8b). In addition to denitrification/anoxic environments, a few studies have observed acesulfame removal under nitrification/aerobic and carbon sources environments [17,29], indicating possible wider adaptation of nitrifying microorganisms and even heterotrophic bacteria for acesulfame biodegradation. A few studies have also observed that biofilm-based processes and higher temperature contributed to acesulfame degradation in WWTP due to higher biomass, versatile microbial communities, and higher microbial activity [5,67,69].

3.5.3. Removal of saccharin, cyclamate, and other sweeteners

Saccharin and cyclamate are easily degradable sweeteners in WWTP in most countries except India and Mozambique, with mean removal efficiency over 90 % globally (Fig. 6b). The relatively low removal of saccharin in India was attributable to insufficient biodegradation[43], while low removal in Mozambique is inconclusive due to limited data points (Fig. 6b). The superior removal is due to the simple biodegradable structures of benzene ring in saccharin and N-cyclohexyl substituent in cyclamate (Table 1), which could be easily broken down through enzymatic reactions in biological treatment [11,9]. Saccharin and cyclamate can be efficiently degraded by nitrifying microorganisms and heterotrophs along with various non-specific oxidative enzymes in WWTP [74]. Saccharin can be degraded into catechol in a two-step process involving the enzyme saccharin dioxygenase by an aerobic bacterium *Sphingomonas xenophaga* SKN [62] (Fig. 8c). Cyclamate is degraded into cyclohexylamine and sulfate by the enzyme cyclamate sulfamatase, while cyclohexylamine will be effectively removed in WWTP (Fig. 8d) [11,33,53]. Aerobic processes clearly outperformed the anaerobic process (i.e. UASB) in saccharin removal, while cyclamate removal was efficient in all wastewater processes (Fig. 6a). No clear temporal differences were found in the removal of saccharin and cyclamate (Fig. 7).

Since influent and effluent data of aspartame, neotame, stevia, and NHDC were insufficient, their removal efficiency in wastewater treatment was not included in this review. Previous studies have observed that aspartame and NHDC were readily removed by both sorption and biodegradation in nitrifying activated sludge [71,74]. Besides, according to the limited available studies, aspartame, neotame, stevia, and NHDC were mostly detected below the LOD, indicating less challenge in WWTP treatment [18,19,66].

4. Discussion

To the best of our understanding, this is the first systematic review to comprehensively analyze the global distribution of artificial sweeteners in influent/effluent and their removal status in WWTP. 3435 documents were retrieved, and 108 documents were included across 24 countries on 6 continents. Sucralose, acesulfame, saccharin, and cyclamate were identified as the most prevalent artificial sweeteners in global wastewater, with a concentration of 0.6–303.0 μg/L in influent and 0.1–81.2 μg/L in effluent (Fig. 2).

Biological treatment processes were revealed to play an important role in the removal of artificial sweeteners (Fig. 6a). CASS, SBR, and wastewater lagoon performed well in sucralose removal due to diverse microbial communities or the prolonged HRT [19,77]. Aerobic processes outperformed the anaerobic process in acesulfame and saccharin removal, and biofilm-based processes also contributed to acesulfame degradation due to diverse microorganisms [22,5,67]. Although saccharin and cyclamate are well removed in current biological processes, sucralose and acesulfame still persist in wastewater treatment. Thus, future studies are encouraged to use novel biological processes to improve the biodegradation of sucralose and acesulfame, such as membrane-aerated biofilm systems and algae-bacteria synergistic systems [39–41,73], which can simultaneously provide aerobic and anoxic environments and versatile microbial communities.

In recent years, increasing trends have been observed in the removal rate of persistent artificial sweeteners (i.e., acesulfame and sucralose) in WWTP. The removal of acesulfame in WWTP increased from −11.8 % (IQR: −27.3 −-5.4 %) during 2008–2011 to 93.5 % (IQR: 83.4–95.5 %) in 2021 globally (Fig. 7). Such improvement in degradation is due to the evolution of denitrifying microbial communities and possible utilization of acesulfame as one of carbon sources for denitrification [28,42,43,7]. Similarly, an increasing trend of sucralose removal in WWTP has been observed worldwide in recent years, especially in China (Fig. 7). Globally, sucralose removal ratio increased from −3.0 % (IQR: −20–9.9 %) before 2017 to 31.3 % (IQR: 6.0–56.7 %) in 2022. In China, sucralose removal ratios in WWTP majorly using A/A/O process, increased from −25.4 % (IQR: −35.4 −-19.8 %) in 2016 to 36.4 % (IQR: 26.7–52.6 %) in 2022 (Fig. S6). In recent years, biodegradation of sucralose has been reported in integrated biological processes with aerobic and anaerobic/anoxic environments (e.g., SBR and CASS process) [16,19]. The improved removal rates may indicate microbial adaptation or evolution for sucralose degradation in WWTP, as occurred in acesulfame removal. Despite this progress, these increased removal rates have been overlooked in previous reviews, and the mechanisms driving this improvement remain unexplored. Thus, future research should focus on

consistently monitoring concentration variations of persistent artificial sweeteners and microbial evolution/pathways for their removal in WWTP, particularly the specific microbial community differences in the WWTP exhibiting high sucralose removal (e.g., WWTP in China).

Sucralose and acesulfame are the major artificial sweeteners (1.1–35.9 μg/L) in effluent worldwide due to their persistence in WWTP, followed by saccharin and cyclamate (mostly <1.0 μg/L) (Fig. 2b). But saccharin and cyclamate are still prevailing in effluent in some developing regions lacking sufficient wastewater treatment capability/facilities (e.g., 81.2 μg saccharin/L in effluent in India) (Fig. 2b). To date, there is no specific regulation for artificial sweeteners in effluent around the world, unlike the increasing number of targeted regulations for emerging pollutants per- and polyfluoroalkyl substances (PFAS) [92]. However, growing evidence has indicated the potential environmental and human hazards posed by artificial sweeteners from effluent on subsequent ecosystems, from the receiving water to wider environments. For instance, higher concentrations (0.1–120.0 μg/L) of artificial sweeteners have been observed in receiving water than the common groundwater (0.01–77.0 μg/L) [78]. Meanwhile, sucralose at 0.05–116.5 μg/L was revealed to elicit the embryonic malformation of zebrafish [10]. High level of saccharin was found to be neurotoxic in *Danio rerio* [20]. Moreover, exposure to artificial sweeteners in environment brings concerns about antibiotic resistance genes (ARG). A recent study revealed that artificial sweeteners stimulated horizontal transfer of ARG in human pathogen [89]. Under the exposure of acesulfame, the number of ARG classes and the abundance of ARG were revealed to increase during anaerobic digestion [85], such ARG risk could deteriorate sludge digestion performance and further spread to wider environments [45,80–83]. Artificial sweeteners in aquatic environments can eventually end up in drinking water, leading to unintended human exposure [47,75,78]. The WHO has reported that long-term use of artificial sweeteners increases the risk of type-2 diabetes, cardiovascular diseases, and mortality in adults [58]. Therefore, it is crucial to closely monitor the presence of artificial sweeteners in WWTP effluent, receiving waters, and drinking water, as well as their impacts on both environment and human health. Future research should also emphasize timely policymaking and regulations, such as setting effluent concentration limits for artificial sweeteners.

Our study also observed inequality in research attention for artificial sweeteners in WWTP, with most studies focused on developed or higher-income countries like North America (28 documents) and Europe (40 documents). Developing countries, such as India (2 documents) [3,70], Brazil (2 documents) [13,2], and Mozambique (1 document) [72], have received limited attention despite consistently reporting higher levels of artificial sweeteners in WWTP. In particular, considering the rapid population growth in developing countries, concentrations of artificial sweeteners in wastewater may increase in these regions in the future. Such inequality might be due to their lack/limited analytical capability in some developing countries. Thus, future research is encouraged to give more attention to developing countries/regions and potential collaborative efforts between developing and developed countries.

In addition, microbial communities and specific enzymes participating in sweetener degradation have been discovered in current research, especially the degradation pathways of saccharin, cyclamate, and acesulfame (Fig. 8). However, the biodegradation of artificial sweeteners from genetic perspective has been limitedly explored. Thus, future research is encouraged to use metagenomic technology to reveal the relationship between enzymatic degradation and microbial community degradation at genetic level.

There are some limitations in this study. First, this systematic review was restricted to only English language documents while the data of some artificial sweeteners might be uploaded/published in local language documents and databases in different countries (e.g. Chinese CNKI, French Persée, and Russian CyberLeninka). Second, due to the lack of systematic and global monitoring of artificial sweeteners, the analysis relied on data reported in existing studies, which often provide time-specific measurements of artificial sweetener concentrations at certain dates and locations. Thus, these data sometimes may not fully reflect the artificial sweetener conditions in countries with insufficient existing studies. Also, seasonal variations in the removal of artificial sweeteners in WWTP were not analyzed due to the lack of sufficient data. Seasonal factors (e.g. temperature, influent loads, etc.) were found to affect sweetener removal performances [90,91]. Thus, future studies are encouraged to pay more attention to seasonal monitoring and removal variations of artificial sweeteners in WWTP. Only a few documents provided the specific operational parameters in wastewater treatment processes, thus the detailed impact of each treatment process and its corresponding operational parameters is not clear. Future studies are highly recommended to conduct through evaluation under controlled conditions to identify the impact of operational parameters on the removal of artificial sweeteners.

5. Conclusions

This systematic review comprehensively evaluated the spatial and temporal distributions of artificial sweeteners in influent and effluent, as well as their removal. Major findings are as follows:

- Sucralose, acesulfame, saccharin, and cyclamate were prevalent artificial sweeteners in influent of WWTP, with a concentration ranging from 0.2 to 303.0 μg/L. The highest concentrations of sucralose, acesulfame, saccharin, and cyclamate were observed in the USA (34.4 μg/L), Spain (58.1 μg/L), India (303.0 μg/L), and Germany (105.3 μg/L), respectively.
- Summer influent generally contained 11.1–33.3 % higher concentrations of artificial sweeteners than other seasons in most countries, while China showed higher concentrations in winter.
- Saccharin and cyclamate are the most easily degradable sweeteners (>90.0 %) in most countries, followed by acesulfame (25.0–70.1 %) and sucralose (-10.0–10.0 %).
- Increasing trends of removal ratios in acesulfame and sucralose have been observed worldwide over time, reaching 93.5 % and 31.3 % respectively in 2022.
- Future research should focus on continuous monitoring of persistent artificial sweeteners in wastewater, novel biological processes, microbial evolution/pathways for sweetener removal in WWTP, timely policymaking/regulations, and developing regions.

Environmental implication

The widespread use of artificial sweeteners has led to their continuous release into wastewater treatment plants (WWTP), raising concerns about the potential impacts of persistent artificial sweeteners on subsequent aquatic ecosystems and human health. Different artificial sweeteners may exhibit different spatial and temporal distributions in WWTPs worldwide. Understanding the global spatiotemporal patterns of artificial sweetener and their removal status in WWTP is vital for improving wastewater treatment capability and evaluating the evolving microbial degradation in WWTP. Advancing treatment technologies and establishing ongoing monitoring and regulatory measures are essential to mitigate the environmental risk of artificial sweeteners.

CRediT authorship contribution statement

Yi Li: Data curation. **Xuan Li:** Writing – review & editing, Supervision, Methodology, Funding acquisition. **Jibin Li:** Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Qilin Wang:** Writing – review & editing, Validation, Supervision, Project administration, Funding acquisition. **Huan Liu:** Resources.

Declaration of Competing Interest

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

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2025.138644.

Data Availability

Data will be made available on request.

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