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Qualitative analysis on contaminants of emerging concern in Swedish landfill leachates: a snapshot of occurrence and spatio-temporal variability

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ABSTRACT

Landfill leachate is a complex and understudied matrix increasingly recognized as a source of contaminants of emerging concern (CECs) to the environment. This study provides a snapshot of the occurrence of CECs in Swedish landfill leachates, investigating their spatial distribution across five landfills, and their temporal pattern over a one-year period at one site. To our knowledge, this is the first study using qualitative non-target and suspect screening to characterize the spatio-temporal profile of CECs in landfill leachate. In total, 79 CECs were identified, including industrial chemicals (ICs), pharmaceuticals and personal care products (PPCPs), per- and polyfluoroalkyl substances (PFASs), pesticides, stimulants, and sweetener. Several compounds, such as triiso-propyl phosphate, 1,3-diphenylguanidine, and 1,3-di-o-tolylguanidine, were reported for the first time in landfill leachate. Spatial analysis revealed a consistent presence of CECs across all sites, with 34 compounds detected in both untreated and treated leachates, indicating limited removal efficiency of existing treatment systems. Temporal monitoring at one landfill showed moderate variation, with the highest number of CECs detected in the winter samples. Persistent detection of ICs, PFASs, and pesticides at all sampling time points suggests continuous leaching and highlights the limitations of current treatment approaches. This study offers important insights into the chemical composition of Swedish landfill leachate and underscores the need for improved monitoring and treatment strategies to mitigate environmental risks associated with CECs.

1. Introduction

Landfill is a common management method for solid waste which cannot be recycled (Siddiqua et al., 2022). Landfills can generate leachate through various pathways: the biodegradation of organic waste, inherent moisture within the waste, and percolation of precipitation through the landfill mass (Nika et al., 2023). Landfill leachate is known to be an important point source of contaminants of emerging concern (CECs) (Siddiqua et al., 2022; Slack et al., 2005; Pisharody et al., 2022; Christensen et al., 2001); and may continue to be released for decades after closure (Propp et al., 2021).

CECs comprise a broad category of chemicals like pharmaceuticals and personal care products (PPCPs), industrial and household chemicals, pesticides, per- and polyfluoroalkyl substances (PFASs), and other unregulated anthropogenic substances, along with their transformation products (TPs) (Tian et al., 2020). These substances are often unregulated; yet many have potential adverse effects on ecosystems and human

health (Nilsen et al., 2019; Feng et al., 2023). For example, some PPCPs have been found to be toxic toward aquatic organisms and linked to endocrine disruption, reproductive toxicity, and carcinogenicity (Cizmas et al., 2015; Wang et al., 2021). Some CECs are classified as persistent, mobile, and toxic (PMT) or very persistent and very mobile (vPvM) substances, which are particularly hazardous due to their longevity and ability to disperse widely in the environment (Malnes et al., 2023).

Due to their extensive use in consumer and industrial applications, CECs often end up in landfills. However, the existing landfill leachate treatment systems are primarily designed to reduce conventional water quality parameters, such as chemical oxygen demand, biological oxygen demand, nutrients, and heavy metals (Nath and Debnath, 2022). As a result, CECs are often insufficiently removed and may therefore be released into the environment by landfill leachate (Masoner et al., 2014; Busch et al., 2010). Therefore, it is important to increase the knowledge about their presence, and thereby the risk for uncontrolled dispersal, in

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landfill leachate.

Numerous studies have reported a diverse array of CECs in landfill leachate using traditional targeted analytical methods, revealing regional variation in the occurrence of CECs. In the United States, pharmaceuticals (e.g., lidocaine, cotinine, amphetamine), household and industrial chemicals such as bisphenol A, camphor, and naphthalene were frequently detected in landfill leachate (Masoner et al., 2014; Masoner et al., 2016). In China, a review pointed out that PPCPs, phthalates, and polycyclic aromatic hydrocarbons (PAHs) were the three most reported groups of CECs in landfill leachate (Qi et al., 2018). Similarly, Swedish studies have highlighted the presence of phenols, phthalates, PAHs, and organic phosphates (Paxéus, 2000; Thörneby et al., 2006; Kalmykova et al., 2013; Kalmykova et al., 2014). These findings demonstrate the widespread occurrence of CECs in landfill leachate worldwide.

However, targeted analysis limits the scope of the investigation, as it focuses only on the pre-selected compounds. To address the challenges of identifying and characterizing a wide spectrum of chemicals, including known and unknown compounds, non-target screening (NTS) and suspect screening (SS) using high-resolution mass spectrometry (HRMS) are often applied. NTS and SS are advanced analytical approaches, which can provide a comprehensive overview of the chemical composition in various environmental samples such as water, sediment, or biota (Hollender et al., 2023; Gonzalez-Gaya et al., 2021). Despite their potential, Nika et al. found only 10 studies worldwide applying NTS and SS to characterize CECs in landfill leachate, most of which were conducted in regions with subtropical or Mediterranean climates, where seasonal variation is relatively mild (Nika et al., 2023). The only study focused in Nordic region (Finland) was based on gas chromatography (GC) - HRMS (Jernberg et al., 2013). However, due to the limitation of GC, the profile of CECs with a wide range of polarity remains largely uninvestigated.

Sweden is an underexplored but valuable case. Its geographical position in the far north of Europe results in pronounced seasonal changes, including wide annual temperature swings and daylight range, variability in precipitation, and freeze-thaw cycles. These environmental conditions can influence the generation and composition of landfill leachate, potentially affecting the presence and abundance of CECs (Yu et al., 2021; Wang et al., 2022). Additionally, Swedish waste management practices before the 21st century were less regulated, allowing some hazardous wastes, such as laboratory residues and medical waste, to be disposed of in landfills (KN departementet, 2001; Avfall Sverige, 2023). There are over 350 active landfills in the country (Swedish EPA, 2022), underscoring the importance of understanding the presence and behavior of CECs in these environments. Furthermore, only a few studies investigated the temporal occurrence pattern of a small number of compounds (Yu et al., 2021; Wang et al., 2022), while most leachate temporal monitoring studies focus on conventional pollution parameters (Kim and Lee, 2009; Mangimbulude et al., 2009; Hoai et al., 2021; Siddiqi et al., 2022).

Despite the concerns about the potential environmental and health impacts of CECs, studies focusing on their occurrence in landfill leachate and the temporal variation are insufficient. This study aims to address this knowledge gap by applying qualitative NTS and SS to capture snapshots of CECs occurrence in selected Swedish landfills. This study is the first to apply a combined NTS and SS methodology to characterize CECs in Swedish landfill leachate, offering a novel spatio-temporal perspective and providing a critical reference point for monitoring efforts in cold-climate regions with similar waste management histories. The findings can enhance the understanding of CECs in landfill leachate and provide a basis for further research on their monitoring, quantification, and development of treatment methods.

2. Methods and materials

2.1. Sampling

To identify CECs in Swedish landfill leachate, two sampling campaigns were independently conducted.

Sampling campaign 1: The aim of this sampling was to identify CECs in Swedish landfill leachate and their spatial distribution across different landfills in the country. Approximately 1.5 L each of untreated leachate (UL) and treated leachate (TL) were grab collected from five selected landfills (A, B, C, D, and E) across Sweden in June and July of 2023 (named individual samples). The selected landfills varied in geographic location, waste composition, size, treatment methods, and age to ensure a representative sample of Swedish landfill leachate.

Sampling campaign 2: The aim of this sampling was to monitor temporal variations in CECs occurrence just in one landfill. Thus, approximately 8.5 L of UL, TL, recipient downstream (RD), and recipient upstream (RU) were collected once per season from landfill B, using 24hour composite time-proportional auto-samplers (Teledyne ISCO, GLS sampler): September 2023 (autumn), January 2024 (winter), March 2024 (spring), and June 2024 (summer). RU samples were collected to establish background conditions in the recipient river and to evaluate the influence of leachate emission. These samples are referred to as seasonal samples. Detailed information on the sampling sites and sampling information is provided in the supporting information section 1 (SI-1). Landfill B was selected to monitor temporal variations due to its representativeness in terms of waste composition, operational practices, and infrastructure. It is a large active landfill receiving mixed waste streams, equipped with a defined leachate collection and treatment system, and provides access for auto-sampling and recipient water monitoring. These features make it a suitable site for investigating temporal patterns in CEC occurrence and leachate treatment performance.

All samples were collected and stored in high-density polyethylene (HDPE) plastic bottles at 10 $^{\circ}C$ during transport and stored at $-22~^{\circ}C$ until analysis.

2.2. Sample preparation and instrumental analysis

Detailed information regarding the chemicals and reagents used in this study are listed in SI-2. All samples regardless of the sampling campaign were prepared equally. Around 600 mL of sample was filtered through glass microfiber filter (Whatman grade GF/F, 0.7 μm pore size, 47 mm in diameter) under vacuum. From the individual samples from sampling campaign 1, two pooled samples were prepared separately alongside the 10 individual samples. The pooled UL was prepared by combining equal volumes (around 120 mL) of filtered UL from each landfill, while the pooled TL was prepared the same way from TL of each landfill (see Fig. S1). The remaining filtered UL and TL from each landfill were kept as individual samples.

Samples were extracted by multi-layer solid-phase extraction (SPE) procedure slightly modified from Gago-Ferrero *et al.* (Gago-Ferrero *et al.*, 2015). Multi-layer SPE cartridges were packed in-house and consisted of a mixed bed containing 100 mg of Sepra ZT-WAX (Phenomenex Strata-X-AW, 115 μm , 330 Å), 100 mg of Sepra ZT-WCX (Phenomenex Strata-X-CW, 100 μm , 300 Å), 150 mg Isolute ENV+ (Biotage, 90 μm , 800 Å) and 200 mg Sepra ZT (Phenomenex Strata-X, 30 μm , 85 Å). In brief, samples were preconcentrated 200 times. Detailed description of the extraction can be found in SI-3.

The analysis was conducted on reversed-phase ultra-high-performance liquid chromatography (Thermo Fisher Scientific, Vanquish) coupled with Orbitrap (Thermo Fisher Scientific, Q Exactive Focus) mass spectrometry (RP-UHPLC-Orbitrap) system, equipped with heated electrospray ionization (HESI) and higher-energy collisional dissociation (HCD). The liquid chromatographic separation system was conducted by Cortecs® C18 (2.1 x 100 mm, 2.7 μ m) column from Waters

(Ireland). The mobile phases consisted of Milli-Q water (Merck, Milli-Q IQ 7000, 18.2 $\mathrm{M}\Omega)$ and methanol (MeOH) (Merck, LC-MS grade). In positive ionization mode, 0.1 % formic acid (Merck) was added to both phases, whereas in negative mode, 5 mM ammonium acetate (Sigma-Aldrich) was used as the additive, both LC-MS grade. The 20-minute gradient elution program was used for both modes. HRMS data were acquired in DDA mode with one full scan MS1 (70 000 resolution) followed by one MS2 scan (17 500 resolution) of the most intense precursor ion, with dynamic exclusion of 3 s.

Detailed instrumental analysis parameters are provided in SI-4 and Table S4. Data analysis was conducted by Compound Discoverer 3.3.200 (Thermo Fisher Scientific).

To minimize contamination and reduce the risk of false positives, all sample containers were pre-cleaned, and handling was conducted using powder-free gloves in clean lab environments. Solvent blanks were analyzed between runs to monitor carryover. NTS and SS feature selection required high confidence in retention time reproducibility, isotope pattern matching, and MS/MS fragmentation, with suspect hits only accepted if they met strict filtering criteria. Compounds detected in blanks were removed from the final data interpretation.

2.3. Non-targeted screening and suspect screening strategy

NTS and SS were applied for qualitative screening of the occurrence of CECs. The detailed NTS and SS workflow can be found in SI-5, and key compound identification parameters are provided in Table S5. The strategy for data processing was to generate an internal suspect list of CECs from the NTS result of pooled samples from sampling campaign 1, and to utilize the suspect list for SS on individual samples and seasonal samples to provide site- and season-specific information. NTS is well known for being laborious and time-consuming and, thus, non-targeted screening of the pooled samples instead of individual samples can improve efficiency drastically. This strategy prioritizes the most abundant and widely occurring compounds across landfills. As for the seasonal samples, NTS was performed to capture the variation in CECs' occurrence among seasons. On top, since the pooled samples were composed of representative landfill leachates in Sweden, the suspect list allows extended screening of CECs in seasonal samples in addition to the NTS.

2.4. Quality assurance and quality control

Quality assurance and quality control strategies consisted of the evaluation of the extraction method performance for a set of 197 CECs (named QC standards, Table S1). QC samples were prepared by spiking 200 mL Milli-Q water with QC standards before the extraction at three concentration levels: 50 ng/L (QC-L), 250 ng/L (QC-M), and 500 ng/L (QC-H). QC samples were extracted in each SPE batch, and the procedural blank (Milli-Q water) was also prepared exactly as the samples for each extraction batch. Procedural blanks (Milli-Q water) were prepared and extracted in parallel with each batch of samples. Compounds detected in any blank at signal intensities >20 % of corresponding sample signals were excluded from further interpretation to minimize the risk of false positives.

Pooled samples and seasonal samples were extracted in triplicate, while individual samples were extracted in duplicate due to sample amount limitations. QC-M sample was injected in between the samples throughout the sequence to ensure the system stability.

Among the 197 QC standards, 158 and 100 of them are detectable in positive and negative modes, respectively. In positive mode, 92 \pm 1 %, 91 \pm 1 %, and 82 \pm 2 % of the spiked QC compounds were detected at the QC-H, QC-M, and QC-L levels, respectively. In negative mode, 95 \pm 0 %, 93 \pm 1 %, and 80 \pm 0 % of QC standards were detected, respectively (Fig. S2). The result of QC samples confirmed the performance of the SPE method in extracting a wide spectrum of CECs from water samples and supported the reliability of the data obtained.

3. Results and discussion

3.1. Identification of contaminants of emerging concern

The combination of both sampling campaigns permitted the identification of 79 CECs in Swedish landfill leachates. Table 1 presents the identified CECs in both sampling campaigns, while additional details are provided in Tables S6 and S7. Among them, 68 were confirmed with reference standards (level 1), and 11 matched with MS/MS library (level 2) (Schymanski et al., 2014). The identified CECs and some of their TPs comprised industrial chemicals (ICs), pharmaceuticals and personal care products (PPCPs), PFASs, pesticides, stimulants, and a sweetener. The distribution of categories of CECs and their level of confidence is visualized as a Sankey diagram (see Fig. 1).

The 31 ICs included 6 phosphate esters, 4 benzotriazoles, 4 benzothiazoles, 2 diphenylguanidines, 2 benzenesulfonamides, and their derivatives, along with some other commonly used chemicals. Phosphate esters, a group of persistent pollutants, are usually used as plasticizers and flame retardants in building materials as well as furniture (Deng et al., 2018; Carlsson et al., 1997), which brings concerns due to their carcinogenicity, reproductive toxicity, and endocrine disruption (Ai et al., 2024). Most of the identified phosphate esters in our study have also been found in landfill leachate in China (Qi et al., 2018; Deng et al., 2018), the US (Masoner et al., 2014; Masoner et al., 2016), Canada (Propp et al., 2021), Norway (Eggen et al., 2010), and Greece (Nika et al., 2020), such as TEP, TBP, and TCEP. However, to the best of our knowledge, TiPP was first detected in landfill leachate in our study, while other studies detected tripropyl phosphate more frequently (Qi et al., 2018). Benzotriazoles are commonly used as corrosion inhibitors, and benzothiazoles are used as rubber vulcanization accelerators (Herrero et al., 2014) and their adverse effects on aquatic organisms and humans were reported in previous studies (Shi et al., 2019; Liao et al., 2018). The BTR, 5-MeBTR, and 2-OHBTH were also found in previous studies on landfill leachate in Greece and the US (Masoner et al., 2016; Nika et al., 2020). In addition, DPG and DTG are commonly used as vulcanization accelerators in rubber production (Li and Kannan, 2024), which are suggested to have health and environmental hazards (Kim et al., 893 (2023)). Their detection in Swedish landfill leachate raises concern, thus, on their potential impact on the recipient environment. To the best of our knowledge, this is the first time they were detected in landfill leachate. The detection of TiPP, DPG, DTG highlights the value of non-target approaches and underscores the strength of non-target screening approaches in uncovering contaminants that may otherwise remain undetected. These findings provide valuable input for environmental risk assessment, particularly by enabling further evaluation of the potential impacts of these compounds on aquatic organisms.

NBBS and NETSA were the two benzenesulfonamides detected in this study, which are used as plasticizers for polyamides, cellulose acetate materials, and other polymer products (Rider et al., 2012; Song et al., 2022). While the neurotoxicity of NBBS and the endocrine-disrupting potential of NETSA have drawn attention (Tian et al., 2020; Eggen et al., 2010), NBBS and NETSA have also been previously found in Norwegian (Eggen et al., 2010), Swedish (Paxéus, 2000; Thörneby et al., 2006), German (Schwarzbauer et al., 2002), and Brazilian (Amaral et al., 2017) landfill leachate. BPA and DMP are two commonly detected endocrine-disrupting compounds (EDCs) related to plastic production, which were found in landfill leachate around the world (Masoner et al., 2014; Qi et al., 2018; Qian et al., 2024). Additionally, BPA is one of the most frequently detected pollutants worldwide (Masoner et al., 2016; Qian et al., 2024).

Twenty-two pharmaceuticals and TPs, and one UV filter were identified in the group of PPCPs, which includes, among others, 6 antihypertensive agents, 5 psychopharmaceuticals and/or anticonvulsants, and 4 analgesic and antipyretic agents. The occurrence of PPCPs can be attributed to the disposal of medicines and cosmetics products through municipal waste (Yu et al., 2021). Toxicological studies have shown the

Table 1The list of identified CECs in Swedish landfill leachates in both sampling campaigns.

ICs			PPCPs		Pesticides	PFASs	
2-NSA	DBHQ	MBTH*	Atenolol acid	Lidocaine	2-Hydroxyatrazine	6:2 FTSA	PFHpS
2OH-BTH	DCHA	NBBS	Bicalutamide	Losartan	Bentazone	FBSA	PFHxA
4-MeBTR	DCU	NBTH	Bisoprolol	Metoprolol	DEET	FHxSA*	PFHxS
5-MeBTR	DMBSA*	NETSA	Candesartan*	Paracetamol	Flamprop-isopropyl*	FOSA	PFNA
8-HQ*	DMBTR	TBP	Carbamazepine	Phenazone	Hexazinone	PFBS	PFOA
BGA	DMP	TBEP	Chlorpropamide	Propyphenazone	Mecoprop*	PFHpA	PFOS
BPA	DPG	TCEP	Desvenlafaxine	Rosuvastatin*	Metalaxyl acid*		PFPeS
BTR	DTG	TCPP	DHB	Sulfapyridine	DMST	Stimulants	Sweetener
BTSA	HMMM	TEP	DiOH-CBZ*	Tramadol	Pyroquilon*	Theobromine	Saccharin
Caprolactam	Laurolactam	TiPP	Fexofenadine	Valsartan		Caffeine	
		TPPO	Lamotrigine	Venlafaxine		Paraxanthine	

Note: Full name and CAS number are provided in Tables S6 and S7. The asterisk (*) indicates the compound was identified as level 2.

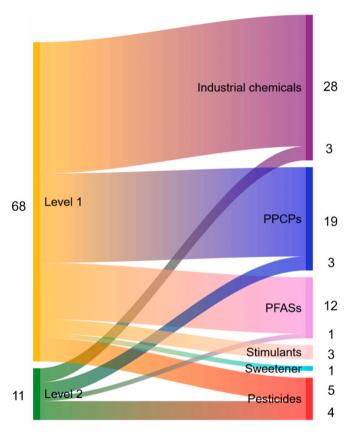


Fig. 1. Sankey diagram of identified CECs in all the landfill leachate samples with the level of confidence. The number of CECs with different levels of confidence from different categories is also shown. Google Charts was used for preparing the figure (licensed under CC BY 4.0 and Apache-2.0).

potentially harmful effects of PPCPs on the aquatic environment and human health (Cizmas et al., 2015; Wang et al., 2021). Bisoprolol, losartan, metoprolol, valsartan, and candesartan are the antihypertensive drugs detected in our study. Metoprolol and valsartan were also detected in landfill leachate in China (Qi et al., 2018), the US (Masoner et al., 2014), and Greece (Nika et al., 2020). Carbamazepine, lamotrigine, and venlafaxine (and its metabolite desvenlafaxine) are the anticonvulsants and/or antidepressant found in this study. Carbamazepine has been intensively investigated and found in many studies around the globe (Qian et al., 2024). Antidepressants, such as venlafaxine and desvenlafaxine, were also detected in the US (Masoner et al., 2014). Paracetamol and tramadol were the analgesic and antipyretic agents identified in our study, and frequently reported in previous studies (Masoner et al., 2014; Masoner et al., 2016; Qian et al., 2024), while propyphenazone was

found in Germany (Schwarzbauer et al., 2002; Schwarzbauer et al., 2006), and Croatia (Matosic et al., 2008; Ahel and Jeličić, 2001). In addition to these compounds, the antihistamine fexofenadine and the anesthetic agent lidocaine have been detected in landfill leachate, particularly in the US (Masoner et al., 2014) and Greece (Nika et al., 2020). Other pharmaceuticals identified in landfill leachate include drugs for hyperlipidemia, antibiotics, antihyperglycemic agents, and antineoplastic agents.

Due to the wide application of PFASs as water repellent, lubricants, and flame retardants, and extensive use of pesticides in agriculture areas, these substances are also the biggest categories of identified CECs in landfill leachate. In our study, 13 PFASs and 9 pesticides were found. Perfluoroalkyl carboxylic acids (PFHxA, PFHpA, PFOA, and PFNA), perfluoroalkyl sulfonic acids (PFBS, PFHxS, PFHpS, PFOS), 6:2 FTSA, and FOSA are 10 PFASs frequently found in landfill leachate (Busch et al., 2010; Qian et al., 2024; Tang et al., 2024). PFPeS, FBSA, and FHxSA are seldom detected in other landfill leachate samples. Regarding pesticides and their TPs, 5 herbicides, 3 fungicides, and 1 insect repellent were identified. The possible sources of pesticides in landfill leachate include the disposal of agricultural waste and being discarded with general waste (Wang et al., 2020). Notably, some of the detected pesticides (e.g., flamprop-isopropyl and hexazinone) have been banned or discontinued in Sweden for decades (Kemikalieinspektionen, 2021; EU, 2022), suggesting their environmental persistence and the potential for long-term leaching from landfill sites. DEET was frequently detected in landfill leachate (Qi et al., 2018; Qian et al., 2024). Herbicides, mecoprop and bentazone, are also detected in landfill leachate in Greece (Nika et al., 2020).

3.2. The spatial distribution of the CECs among five landfills

This section aims to compare the occurrence and composition of CECs across five Swedish landfills, based on a single summer sampling event, and to explore how site-specific factors (e.g., waste input, leachate treatment, operational practices) may explain observed differences. CECs' occurrence frequency and their spatial distribution were investigated during the first sampling campaign. In total, 76 CECs were identified across all 5 landfills, including 31 ICs, 22 PPCPs, 10 PFASs, and 9 pesticides, among others. Fig. 2 presents the occurrence of CECs in untreated (UL) and treated (TL) leachates of in each landfill. A total of 43 contaminants of CECs were detected in the UL of all five landfills examined. Of these, 34 CECs were also present in TL of all landfills, indicating that current treatment methods are insufficient in effectively removing these substances from landfill leachate. The compounds that were detected in UL and TL of all landfills were grouped into CECs set 1 in order to simplify the visualization (see Fig. 2).

Twenty-eight ICs were frequently detected in at least four out of the five landfills. This group included a range of compounds such as flame retardants, plasticizers, rubber accelerators or crosslinking agents, corrosion inhibitors, and agents used in chemical or fiber synthesis. The

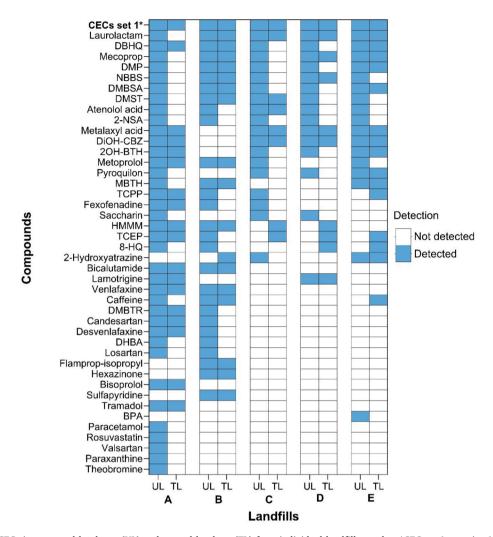


Fig. 2. Occurrence of CECs in untreated leachates (UL) and treated leachate (TL) from individual landfill samples. *CEC set 1 contains 17 ICs,10 PFASs, 5 pharmaceuticals, and 2 pesticides. Please refer to SI-7 to have the full list.

consistent presence of these compounds across multiple sites suggests a common source or widespread use of these substances in the region.

Pharmaceuticals also showed a notable presence, with eight compounds, including antihypertensive agents, anticonvulsants, and analgesic agents, frequently detected in at least four out of five landfills. This pattern highlights the persistent nature of pharmaceutical residues in waste streams entering Swedish landfills. PFASs were detected in all landfills, with all 10 PFASs identified in the first sampling campaign present at each site, and six out of nine pesticides analyzed were detected in at least four landfills.

The widespread occurrence and consistent detection of CECs across multiple landfills suggest a significant and uniform contamination pattern throughout Sweden. The absence of distinct spatial distribution characteristics implies that these contaminants may stem from widespread usage and disposal practices rather than localized sources. This uniformity underscores the urgent need to improve current treatment technologies to effectively address the removal of these persistent contaminants from landfill leachate.

While many CECs were widely detected in selected landfills, the occurrence of others varied among sites. A total of 19 CECs presented only in at most 2 landfills in the sampling campaign 1, including 13 PPCPs, 2 pesticides, 2 ICs, and 2 stimulants.

Among the 15 PPCPs and stimulants, 7 were found exclusively in landfill A, while 6 were found in both landfills A and B. The differences in the occurrence of PPCPs and stimulants may result from several

factors. Both landfills A and B received sludge from wastewater treatment plants (WWTPs), where some compounds are retained (Yu et al., 2024), while others might have been directly disposed of in the landfills. The higher detection frequency of certain pharmaceuticals may reflect their widespread use (Wang et al., 2020; Richardson and Kimura, 2020). Although the presence of PPCPs is commonly attributed to the disposal of unused drugs in household waste (Yu et al., 2021), it should also be noted that sewage sludge mixed with other materials is often used as a protective layer on top of the cap of landfill in Sweden (Sundberg et al., 2003; Statistikmyndigheten, 2024), which can contribute to the occurrence of PPCPs in landfill leachate (Pérez-Lemus et al., 2019).

As for pesticides, herbicides hexazinone and flamprop-isopropyl were both only found in landfill B. The limited detection of flamprop-isopropyl may be attributed to its specific application in controlling wild oats in wheat and barley, limiting its use in areas cultivating these crops (Kemikalieinspektionen, 2021).

DMBTR and BPA are the ICs that were only found in landfills A and B, and landfill E, respectively. DMBTR was frequently detected in water samples, but previous review suggested its concentration was often found lower than other benzotriazoles (Shi et al., 2019). One possible explanation is that DMBTR seems to be relatively easy to be biotransformed (Shi et al., 2019). In several studies, BPA was the most frequently detected CEC in landfill leachate (Masoner et al., 2014; Masoner et al., 2016; Nika et al., 2020; Qian et al., 2024), but it was detected in only one landfill in this study. That might be attributed to the

restricted use of BPA at multiple levels in the EU and Sweden (Udovyk, 2014).

The difference in the CECs' occurrence between UL and TL among different landfills was also investigated. Across all five landfills, 76 CECs were identified in UL samples and 66 in TL samples, suggesting a reduction in the number of detected CECs after treatment. Masoner et al. also observed the higher total number of detected CECs in UL compared with TL in a study conducted in the US (Masoner et al., 2016). However, in our study the majority of the CECs detected in UL remained detectable after the treatment, indicating that current treatment processes may have limited impact on the removal of many CECs from landfill leachate. The difference in treatment methods among landfills may explain the variation in observed CECs occurrence between UL and TL samples. In landfill A, the leachate was treated by multiple aerated wetland ponds followed by soil infiltration. The wetland has been shown to have relatively good removal efficiencies (Bakhshoodeh et al., 2020), while the aeration process could further improve the treatment performance (Nivala et al., 2007). In addition, infiltrating the preliminarily treated leachate through the soil field might further remove the CECs with soil and plant adsorption, and microbial biochemical reactions (Wang et al., 2025; Zhang et al., 2007). Conversely, most of the CECs found in UL were also present in TL at landfill E, where the leachate was only pretreated using aerated ponds before transferring the leachate to a local WWTP.

Some CECs were detected in TL but not in their corresponding UL, such as TCEP, HMMM, 8-HQ, and TCPP, likely due to the timing of the sampling events and the setup of the landfill facilities. Whereas the UL and TL samples of each landfill were collected on the same day in the sampling campaign 1, the leachate hydraulic retention time during the treatment process could take up to days or even weeks (Renou et al., 2008; Kurniawan et al., 2010), resulting in the collected TL potentially originating from an earlier batch of leachate than the corresponding UL sample. Besides, leachate composition may be influenced by other inputs from onsite activities. As observed during sample collection, several investigated landfills have some sectors to receive, temporarily store, and process contaminated soil, wood materials, and other waste without landfilling. The surface runoff from these sectors could flush into the treatment chain and, thus, change the composition of leachate and contribute to the CECs in sample periodically. Potentially, such input could lead to a short-term spike in CECs' load in UL and a prolonged impact on the treatment chain due to the extended process time. Sampling at both the inlet and outlet within a single day did not account for the hydraulic retention time of the leachate treatment process, potentially leading to non-detection of CECs at the inlet while detecting them at the outlet. The intermittent and unpredictable nature of external inputs to the leachate stream introduces additional variability, further complicating the characterization of leachate composition. It should be noted that the spatial sampling was limited to a single time point (summer 2023), and CEC concentrations may vary across seasons due to climatic and operational factors. Thus, the spatial differences reported here represent a snapshot under summer conditions, and further multiseason sampling is needed to confirm whether these patterns persist vear-round.

In summary, spatial analysis revealed a core set of CECs consistently present across all landfills, suggesting common waste sources and widespread use of certain compounds (e.g., PFASs, phosphate esters, common pharmaceuticals). However, distinct profiles were observed in some landfills—for instance, Landfill A showed higher frequencies of PPCPs, likely due to past acceptance of sewage sludge and medical waste. Differences in leachate treatment methods (e.g., wetlands, soil infiltration) also appear to influence removal patterns. Overall, these findings highlight both shared and site-specific characteristics in the spatial occurrence of CECs in Swedish landfill leachate.

3.3. Snapshot of temporal variations in CECs in one landfill

Environmental factors such as precipitation, temperature, and sunshine duration may influence the occurrence of CECs in landfill leachate through the impact on mobilization, dilution, degradation and treatment performance (Yu et al., 2021; Wang et al., 2022). To capture a temporal snapshot of CECs presence under varying seasonal conditions, 24-hour composite samples were collected once per season over a one-year period at four sampling locations—UL, TL, RD, and RU—at land-fill B. Across four seasons, a total of 70 distinct CECs were detected, including 29 ICs, 18 PPCPs, 13 PFASs, 8 pesticides, and 2 stimulants. Detailed information on the detection of each CEC at all locations is presented in Fig. S3.

Fig. 3 presents the total number of detected CECs across all locations, illustrating the temporal trends in their occurrence. Overall, the total number of detected CECs varied moderately by season, with winter exhibiting the highest (67) and summer showing the lowest total number (56) of detected CECs across all locations with ICs remaining the dominant category in all seasons. The second largest category shifted from PPCPs in autumn and winter to PFASs in spring and summer, as the number of detected PPCPs declined in spring and summer. Among the five CEC categories, PPCPs showed the most pronounced temporal variation. These changes indicate that some CECs showed notable fluctuations while the majority of them remained relatively consistent throughout the year.

To further characterize the temporal distribution of individual CECs, Fig. 4 summarizes their occurrence at UL, TL, and RD. RU data was excluded due to low detection frequencies to enhance the focus on landfill-related contaminants.

Around two-thirds of the detected CECs were present at all three locations throughout all seasons. These CECs are referred to as CECs set 2 in Fig. 4 to simplify the visualization. The CECs set 2 included most of the ICs (22/29), PFASs (11/12), and pesticides (7/8) identified in sampling campaign 2. Their year-round consistent detection highlights their persistence and suggests limitations in current treatment technologies.

Winter not only had the highest total number of detected CECs, but the highest number of CECs detected at all three locations as well. In comparison, autumn and spring had fewer detected CECs and showed lower detection frequencies among locations, while summer had the fewest detected CECs. Autumn recorded the highest number of CECs at a single location, with 14 CECs detected only in UL. Winter and spring had the highest numbers of CECs detected in only two locations: six CECs were found at both UL and TL in winter, while six CECs were detected exclusively at TL and RD in spring. The observed variation is likely

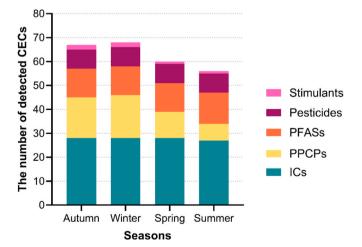


Fig. 3. The number of detected CECs by different categories in four seasons across all locations at landfill B.

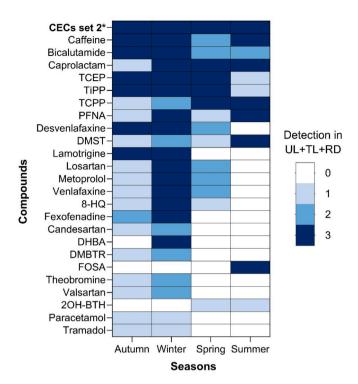


Fig. 4. The occurrence of CECs in untreated leachate (UL), treated leachate (TL), and recipient downstream (RD) across four seasons at landfill B. Colored boxes indicate detection frequency, representing the number of locations in which each compound was detected. *CECs set 2 contains 46 compounds, including 22 ICs, 11 PFASs, 7 pesticides, 6 pharmaceuticals. Please refer to SI-7 to have the full list.

attributed to various combined factors, including precipitation, temperature, sunshine duration, and operational factors. The detailed precipitation and temperature data are shown in Fig. S4.

Previous studies pointed out precipitation can affect the production of the leachate (Tränkler et al., 2005) and the amount of organic matter in the landfill leachate (Kim and Lee, 2009; Mangimbulude et al., 2009; Hoai et al., 2021; Siddiqi et al., 2022). Moderate precipitation can mobilize the CECs from waste into leachate and therefore increase the abundance of CECs (Masoner et al., 2014; Yu et al., 2021), while heavy precipitation may dilute the leachate and lower the abundance of CECs (Wang et al., 2022; Lu et al., 2016). During the winter sampling event, heavy snowfall followed by snowmelt likely mobilized CECs from waste, while also increasing the flow in the recipient river, potentially diluting downstream abundance. This may explain detection of some CECs in UL and TL but not in RD. In contrast, spring and summer sampling events conducted during the period with low precipitation, which may account for non-detections in UL. In autumn, a two-week dry period followed by intense rainfall one day before sampling (Fig. S4) may have led to a sudden release of accumulated CECs into leachate (Yu et al., 2021). However, the leachate takes days or weeks to go through the treatment chain, so the CECs flushed into the UL during rainfall may not yet reach the TL and RD sampling points.

Precipitation can also affect the hydraulic retention time in the treatment system. According to the non-public leachate flow records from landfill B, increased flow during winter and spring likely shortened retention time in the treatment system, potentially limiting the extent of contaminant removal (Albornoz et al., 2020; Toet et al., 2005; Majewsky et al., 2011). This may explain the higher number of CECs detected in TL and RD during these seasons compared to summer and autumn.

Temperature can be another crucial factor contributing to the temporal variation of CECs. Lower temperature in winter and spring could influence the microbial activity in both landfill waste (Wang et al.,

2012) and the treatment system (Sui et al., 2011; Kadlec and Reddy, 2001), potentially contributing to the higher number of detected CECs in TL and RD during these seasons. Combined with the factor of precipitation mentioned above, some CECs might be degraded in the waste before released into leachate, potentially explaining low detection in UL in summer. Additionally, seasonal differences in sunshine duration may also play a role. Prolonged sunshine duration and elevated temperature may facilitate the phytoremediation in polishing ponds through increased vegetation growth (Jones et al., 2006) and enhance photodegradation (Andreozzi et al., 2003), further contributing to lower CECs detection during these seasons.

Furthermore, as mentioned in section 3.2, the surface runoff from the waste storage sectors, the timing of waste intake, and the sampling setup could introduce additional variability affecting CEC detection. These findings underscore the importance of further investigation to fully reveal the relationship between operational and seasonal factors and CECs' occurrence.

4. Conclusion

This study provides a snapshot-based but comprehensive overview of CECs in Swedish landfill leachate, highlighting their occurrence, spatial distribution, and temporal variability. It is the first study to investigate spatio-temporal CECs profile using qualitative NTS and SS by UHPLC-HRMS. Across two sampling campaigns, a total of 79 CECs were identified, including ICs, PPCPs, PFASs, pesticides, stimulants, and a sweetener. Many compounds—such as triisopropyl phosphate, DPG, and DTG—were detected in landfill leachate for the first time.

CECs' occurrence frequency and their spatial distribution were investigated during the first sampling campaign. In total, 76 CECs were detected across five studied landfills, with 43 found in all sites, implying that homogenized distribution of these contaminants may contribute to widespread usage and common disposal practices rather than localized sources. The snapshot provided by this study reveals that current landfill leachate treatment methods are insufficient for removing a substantial portion of CECs, as many were detected in both untreated and treated leachates. The temporal variability was studied in one landfill and our results have shown moderate fluctuations in CEC occurrence, with winter presenting the highest burden (68 CECs were detected)—likely due to a combination of precipitation, temperature, retention time, and operational factors. Although temporal trends were studied at a single site, Landfill B is operationally and climatically representative of many active landfills in Sweden. Thus, the insights gained may serve as a useful reference for similar Nordic and cold-climate landfill systems.

These findings underscore the need for improved treatment technologies targeting persistent CECs. While the study offers important insights, it also highlights the necessity of long-term monitoring to capture temporal dynamics more robustly. Our results can enhance the understanding of CECs in landfill leachate and provide a basis for further research on their quantification and development of treatment methods.

CRediT authorship contribution statement

Tsz Yung (Patrick) Wong: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Writing – original draft. Alberto Celma: Writing – review & editing, Investigation, Data curation, Conceptualization. Natalie Storm: Methodology, Investigation, Formal analysis, Data curation. Malin Hultberg: Writing – review & editing, Conceptualization. Oksana Golovko: Writing – review & editing, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

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. Supplementary data

Supplementary data to this article can be found online at $\frac{https:}{doi.}$ org/10.1016/j.envint.2025.109834.

Data availability

Data will be made available on request.

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