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# A review of properties, occurrence, fate, and transportation mechanisms of contaminants of emerging concern in sewage sludge, biosolids, and soils: recent advances and future trends

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Treated sewage sludge, or biosolids, are used worldwide to improve soil quality and fertility due to their high organic matter content and micro- and macronutrients. However, biosolids also introduce contaminants into the environment. This review assessed recent publications on the occurrence, environmental fate, and transportation mechanisms of 419 contaminants of emerging concern (CEC) across four matrices: sewage sludge, biosolids, soils, and dust. Among these, 229 CECs were positively detected at least once. The review focuses on various CECs, including 18 therapeutical classes of pharmaceutical products, personal care products (parabens and chlorophenolic antimicrobials), hormones, plastic-related compounds (bisphenols and phthalates), rubber antioxidants (substituted diphenylamines and para-phenylenediamines), and neonicotinoid insecticides. Phthalates dominate biosolid composition, accounting for over 97% of the total weight of CECs investigated, followed by pharmaceuticals (1.87%), personal care products (0.57%), hormones (0.09%), antioxidants (0.07%), and bisphenols (0.05%). The 50 most prevalent contaminants include phthalates [e.g., di (2ethylhexyl) phthalate-DEHP, butyl benzyl phthalate-BBzP], antifungals (e.g., miconazole-MCZ, ketoconazole-KTZ), chlorophenolic antimicrobials (e.g., triclocarban-TCC, triclosan-TCS), fluoroquinolone antibiotics (e.g., ofloxacin-OFL, ciprofloxacin-CPF), cardiovascular medications (e.g., telmisartan-TMS, propafenone-PPF), analgesics (e.g., salicylic acid-SA, naproxen-NPX), hormones (e.g., mestranol-EEME, progesterone-P), antidepressants (e.g., sertraline-SRT, amitriptyline-AMT), and lipid regulators (e.g., fenofibrate-FNF). Analytical techniques like matrix-assisted laser desorption/ionization mass spectrometry imaging (MALDI-MSI) have emerged as a valuable tool for qualitative analysis, allowing for identifying CECs in biosolids. The environmental fate and transport dynamics of studied CECs are influenced by their physicochemical properties (water solubility, volatility, degradation time, sorption capacity, and bioaccumulation potential) and environmental conditions (temperature, pH, and moisture content). Furthermore, soil characteristics, such as composition, organic matter, and microbial activity, play key roles in their adsorption, degradation, and persistence in soil environments. Additionally,

wastewater treatment processes impact the transformation and removal of CECs, affecting their degradation and partitioning between treated effluents and sewage sludge.

KEYWORDS

sewage sludge, biosolids, soil, emerging contaminants, physicochemical properties, occurrence, fate and transport

## **1** Introduction

In recent years, a group of contaminants referred to as emerging contaminants (ECs) or contaminants of emerging concern (CECs) has generated extensive research interest because of their toxic potential for humans and the ecosystem, especially since they are usually not regulated (Sauvé and Desrosiers, 2014). These CECs are ubiquitous because they have been detected in several environmental compartments, starting from simple matrices like air (Cao et al., 2022), drinking water (Teymoorian et al., 2023; Wu et al., 2023), surface water (Goeury et al., 2019; Zhong et al., 2022), moderately dense media exemplified by road dusts (Chae et al., 2024) and suspended sediments (Fenni et al., 2022; Shang et al., 2023), but also including more complex substrates such as manure (Solliec et al., 2016; Zahedi et al., 2022), composts (Huang et al., 2023; Tran et al., 2023), municipal wastewaters (Rabii et al., 2014; Vaudreuil et al., 2022), agricultural and urban soils (Cao et al., 2022; Rødland et al., 2023; Zhang et al., 2023), sewage sludge (Boix et al., 2016) and biosolids (Larivière et al., 2017; Pérez-Lemus et al., 2019; Kumar et al., 2022; Munoz et al., 2022; Saliu and Sauvé, 2024).

The sources of CECs encompass various nonpoint and point sources of pollution (Stefanakis and Becker, 2015), including air deposition (Barroso et al., 2019), sweeping runoff (Sidhu et al., 2013), agricultural enrichment, and pesticide application (He et al., 2015), plastic mulching (Hofmann et al., 2023), sediments, soils (vadose and phreatic zones), surface water bodies (rivers, lakes, seas, oceans), groundwater, drinking water, wetlands, humans, and biota (Ashraf et al., 2023). Additional documented sources are landfill sites, septic tanks (Ashraf et al., 2023), domestic and industrial discharges (Smith, 2009), hospital outflows (Vaudreuil et al., 2022), wastewater treatment plants (WWTP) influents and effluents (Rout et al., 2021; Vaudreuil et al., 2022; Ashraf et al., 2023), as well as soil manure and biosolids applications (Hanselman et al., 2003; He et al., 2015; Larivière et al., 2017; Pérez-Lemus et al., 2019).

Biosolids represent nutrient-rich final product of sewage sludge stabilization, achieved through various wastewater techniques including composting, aerobic or anaerobic digestion, lime treatment, or dewatering techniques such as air drying, vacuum filtering, and centrifugation. This process yields an organic solid product that is deemed safe for agricultural applications (Zorpas and Inglezakis, 2012). The United States Environmental Protection Agency (USEPA) categorizes biosolids into two distinct classes based on their pathogen content: Class A, which comprises biosolids that are virtually free of pathogens and can be applied without any sitespecific restrictions, and Class B, which may harbor pathogens and thus mandates the implementation of management practices and site limitations to safeguard public health (USEPA, 1994). In Europe, the regulations pertaining to soil protection advocate for the treatment of sewage sludge prior to its application in agricultural practices, without designating it as biosolids (European Communities, 1986).

While the meticulous application of biosolids for soil quality and fertility improvement has been documented in California since 1920 and in Japan since 1950 (Kalavrouziotis et al., 2023), their regulation and control have garnered substantial government attention in the not-so-distant past (USEPA, 1994) given the risks to the ecological and public health that they pose (Girovich, 1996; McBride, 1995; Mannina et al., 2023). From an agricultural stance, amending soils with biosolids supplements organic matter (Zhang, 2017; Kumar et al., 2022; EPA, 2023), macro- (e.g., N, P, S, Ca, Mg) and micro-nutrients (e.g., Mn, Cu, Zn, Mo, K) to cultivable lands (Kim and Owens, 2010; Wijesekara et al., 2016). In contrast, these substances harbor a concentrated presence of contaminants that have adsorbed onto the soil post-wastewater treatment (Ashraf et al., 2023). The chemical composition and characteristics of biosolids (processed sewage) are widely variable and largely influenced by various factors such as their origin, treatment techniques, age, moisture levels, and temperature (Wijesekara et al., 2016; Wang et al., 2009; Zhang, 2017).

Latest literature reviews on the occurrence of CECs in sewage sludge, biosolids, and soils have mainly concentrated on the assessment of particular classes, namely, pharmaceutical products (PPs) (Larivière et al., 2017), pharmaceutical products and personal care products (PPCPs) (Pérez-Lemus et al., 2019; Riva et al., 2021; Ashraf et al., 2023), phthalates (He et al., 2015; Gao et al., 2018), per-and polyfluoroalkyl substances (PFAS) (Riva et al., 2021; Kumar et al., 2022; Saliu and Sauvé, 2024), endocrine disrupting chemicals (EDCs), surfactants and flame retardants (Kumar et al., 2022), and plastic leachates and particles (Kumar et al., 2022; Hofmann et al., 2023). Others focused on analytical techniques for characterizing them into those complex matrices (Larivière et al., 2017; Martín-Pozo et al., 2021; Kumar et al., 2022), while others aimed at their occurrence, fate, or removal during drinking water and wastewater treatment (Kumar et al., 2022; Yang et al., 2017). This literature review's original contribution is to annotate the occurrence of twenty-seven classes of CECs: 18 therapeutic classes of PPs, two classes of PCPs, natural and synthetic hormones, two classes of plastic-related compounds (bisphenols and phthalates), two classes of rubber and polymer antioxidants (aromatic secondary amines), and neonicotinoid insecticides in complex matrices, namely, sewage sludge, biosolids, soils, and dusts. The review will also present the physicochemical properties of studied CECs, discuss factors determining their behaviors during the wastewater treatment (WWT) process, and debate their environmental fate and transportation mechanisms in soils.

## 2 Materials and methods

To examine the occurrence of specified CECs in aforesaid environmental compartments, a thorough examination of

scientific literature published between January 2018 and July 2023 was conducted across four reputable academic databases (Web of Science, PubMed, Reaxys, and SciFinder). However, a thorough analysis of articles and reports from before and after this timeframe was also conducted to provide a comprehensive elucidation of the characteristics of CECs, their means of mobility, and their eventual destiny in the environment. The review was carried out through cross-referencing search terms organized into three distinct clusters: the types of matrices under investigation (sewage, biosolids, soil), the main groups of targeted CECs, and thematic research areas (occurrence, transport, fate). The search query was tailored to exclude references related specifically to wastewater influents and effluents to enhance precision and clarity in the retrieved citations. After deduplicating the retrieved articles, a thorough examination of their titles and abstracts was conducted, followed by a classification based on thematic relevance. Finally, we surveyed the references within the acquired publications and performed a targeted search on Google Scholar to obtain any missing information. A total of 151 articles were retained for a thorough and detailed review of the occurrence, environmental transport, and transformation processes of target groups of CECs. Figure 1 summarizes the approach for this literature review.

## **3** Pharmaceutical products

### 3.1 Overview

PPs, also known as medicines, medications, or drugs, are preparations administered in conventional and traditional medicine to diagnose, protect (public health), prevent (prophylactic), treat (therapeutic) humans or animals against disease or health conditions, or to restore, correct, and alter organ functions (Daughton and Ternes, 1999; WHO, 2024; Larousse, 2024). PPs are often categorized based on their mechanisms of action or therapeutic applications, and this review examines the presence of PPs across 18 therapeutic classes within sewage sludge and biosolids. These classes are analgesics (narcotic and non-narcotic analgesics, non-steroidal anti-inflammatory drugs-NSAIDs, antipyretics, stimulants, and their metabolites), antibiotics (sulfonamides, quinolones and fluoroquinolones, βlactams, macrolides, tetracyclines, nitroimidazoles, lincosamides, nitrofurans, oxazolidinones, and ionophores), anticonvulsants and antiepileptics, antidepressants (comprising antipsychotics and anxiolytics), cardiac care medications (antiarrhythmics, antiplatelets, antihypertensives, β-blockers, diuretics, calcium channel blockers), antifungals, lipid regulators (statins and fibrates), antihistamines and bronchodilators, antineoplastics (listing alkylating agents, antimetabolites, and immunosuppressants), erectile dysfunction agents, x-ray contrast media, antidiabetics, antivirals, muscles relaxants, anesthetics, cholinergic drugs, antihelminthics, and antiemetics). A complete enumeration of the covered PPs and their respective therapeutic classifications is available in the Supplementary Appendix B: Sheet 2-Analytes.

# 3.2 Physical and chemical characteristics: indices of fate and transport

From a chemical standpoint, PPs are engineered to be moderately water-soluble to facilitate physiological distribution, and to be lipophilic to ensure their bioavailability and biological activity (Ikehata et al., 2006). The physicochemical properties of PPs exhibit considerable variability, with aspects such as structural simplicity seen, for instance, in fluorouracil and metformin



(MTF), versus structural complexity, as perceived in aclarubicin and roxithromycin. Additionally, these characteristics include molecular weight variations, exemplified by MTF having a molecular weight of 129.16 g/mol and tylosin at 916.10 g/mol. Furthermore, the acid dissociation constant (pKa) of PPs fluctuates remarkably, as in the case of oxazepam with a pK<sub>a</sub> of 1.63 (Shayesteh et al., 2015), oxycodone presenting a pKa of 8.5 (Kinnunen et al., 2019), and simvastatin with a pKa of 14.91 (Shaheen et al., 2022). The n-octanol-water partitioning coefficient (log Kow) values also demonstrate notable differences; for example, tetracycline exhibits a log Kow of -1.37 (Chabilan et al., 2022), gemfibrozil (GMF) with a log K<sub>ow</sub> of 4.39 (Pérez et al., 2023), while mefenamic acid displays a log K<sub>ow</sub> of 5.12 (Feng et al., 2013). The presence of more than one functional group in a drug's chemical architecture normally causes it to have more than one pKa value. Gao et al. (2019) proposed a hydrophilicity-hydrophobicity scale for organic contaminants, predicated on the log Kow value. They categorized organic contaminants into hydrophilic (log K<sub>ow</sub> < 3), moderately hydrophobic (3 < log  $K_{ow}$  < 5), and strongly hydrophobic (log  $K_{ow}$  > 5). In parallel, the Stockholm Convention on Persistent Organic Pollutants (POPs) designates a log  $K_{ow}$  value greater than five to indicate a pollutant's potential for bioaccumulation in aquatic organisms (UNEP, 2019). Table 1 presents important physicochemical properties (solubility, volatility, degradation time, sorption, and bioaccumulation) of organic contaminants, predictors of their environmental destiny, and behaviors.

# 3.3 Occurrence in sewage sludge and biosolids

In June 2019, the German Environment Agency (UBA) conducted a comprehensive review of the presence of PPs in various environmental matrices to facilitate an update of the Measured Environmental Concentration Database (MEC DB). That review reported a total of 54 environmental matrices

TABLE 1 Physicochemical properties indicative of the transport mechanism and environmental fate of organic contaminants.

Water solubility (S <sub>w</sub> ) FAO (2000)	Solubility (mg/L at 20°C)	Classification
	<0.1	Not soluble
	0.1–1	Slightly soluble
	1–10	Moderately soluble
	10-100	Readily soluble
	>100	Highly soluble
Henry's Law Volatility Constant ( <i>Volatility-H<sub>v</sub></i> ) Howard (2017) Sander et al. (2022)	Volatility (atm-m <sup>3</sup> /mol)	Classification <sup>a</sup>
	<10 <sup>-3</sup>	Highly volatile
	$10^{-3} - 10^{-7}$	Moderately volatile
	>10 <sup>-7</sup>	Slightly volatile
Biological and physicochemical degradation (Half-life degradation time-DT <sub>50</sub> ) FAO (2000)	DT <sub>50</sub> (days)	Classification
	<20	Readily degradable
	20-60	Fairly degradable
	60–180	Slightly degradable
	>180	Very slightly degradable
Sorption (Organic carbon-water partitioning coefficient-log K <sub>oc</sub> ) FAO (2000)	Log K <sub>oc</sub>	Classification
	<1	Highly mobile
	1–2	Mobile
	2-3	Moderately mobile
	3-4	Slightly mobile
	4–5	Hardly mobile
	>5	Immobile
Bioaccumulation ( <i>n-Octanol-water partitioning coefficient-log K<sub>ow</sub></i> ) Gao et al. (2019) and UNEP (2019)	Log K <sub>ow</sub>	Classification
	<3	Moderately bioaccumulative
	3–5	Bioaccumulative
	>5	Highly bioaccumulative

<sup>a</sup>Scale adjusted in three levels



clustered into liquid emissions (e.g., reclaimed water, hospital sewage), liquid immissions (e.g., drinking water, groundwater), solid emissions (mainly sewage sludge and biosolids), and solid immissions (e.g., soil, lake sediments). The analysis positively (>LOD) determined from seven solid emissions 145 PPs and metabolites across 75 countries (Eike et al., 2019). A subsequent update released in December 2021 broadened this assessment in 89 countries and incorporated an additional category of composts in solid emissions, leading to the identification of 337 PPs from eight solid matrices (Graumnitz and Jungmann, 2021). A comparative analysis between the initial report, which reviewed scientific literature from 2010 to 2016, and the most recent one, which focused on publications from 2017 to 2020, reveals that an additional 192 PPs have been newly and positively identified as originating from solid emissions.

This review encompasses an analysis of academic articles discussing PPs in sewage sludge or biosolids. Moreover, publications about the fate and transport of PPs were examined to clarify their ecological destiny and migration routes during WWT processes and within soil matrices. From a total of 301 PPs and degradation products analyzed, 112 compounds were positively detected at least once in one of the two matrices under consideration. The most concentrated pharmaceuticals are dominated by over-the-counter medicines (antifungals and analgesics), followed by antibiotics and antidepressants. Firstly, miconazole (MCZ: 10,382 ng/g) (Li et al., 2021), followed by naproxen (NPX: 9,355 ng/g) (Pérez-Lemus et al., 2020), ciprofloxacin (CPF: 4,889 ng/g) and ofloxacin (OFL: 4,673 ng/g) (Riva et al., 2021), norfloxacin (NRF: 3,359 ng/g) (Camotti Bastos et al., 2020), ketoconazole (KTZ: 3,009 ng/g) (Svahn and Björklund, 2019), salicylic acid (SA: 2,695 ng/g) (Pérez-Lemus et al., 2020), amitriptyline (AMT: 1,980 ng/g) (Costa Junior et al., 2020), diclofenac (DCF: 1,620 ng/g) (Camotti Bastos et al., 2020), and caffeine (CAF: 1,620 ng/g) (Costa Junior et al., 2020). Figure 2

reveals the mean concentration of the 30 most prevalent PPs in sewage sludge and biosolids. The cumulative mean concentration of various classes of PPs in sewage sludge is primarily dominated by antibiotics (7,689 ng/g), analgesics (4,989 ng/g), antidepressants (2,607 ng/g), cardiovascular medications (1,723 ng/g), and antifungals (1,598 ng/g). Antifungals (8,633 ng/g) are the main class of PPs found in biosolids, followed by cardiovascular agents (3,501 ng/g), antibiotics (1,824 ng/g), antidepressants (1,073 ng/g), and lipid regulators (622 ng/g). The cumulative mean concentration of PPs in sewage sludge (18,949 ng/g) does not substantially differ from that in biosolids (16,324 ng/g). For the summary statistics of various therapeutic classes, refer to Table S1 in Supplementary Appendices A, B-Sheet 3-PPs.

# 3.4 Environmental fate and transportation mechanisms

In the context of WWT, the characteristics of PPs, the concentration of dissolved organic matter (DOM) that enhances their leaching into effluent, and pH levels emerged as critical factors influencing the environmental fate and transport mechanisms of PPs (Zhang et al., 2014; Silva et al., 2021). Despite the limited availability of treatment technologies specifically addressing PPs and the inadequacy of standard WWT processes in effectively eliminating them, various advanced treatment methods have significantly improved the removal of pharmaceutical compounds during WWT. They include conventional activated sludge, moving bed biofilm reactors (MBBR) (Tisler et al., 2021), ultraviolet C (UV-C) radiation (Grgić et al., 2021), advanced oxidation such as electrochemical oxidation, the use of microalgae and fungal strains, anaerobic membrane bioreactors, as well as membrane separation techniques including nanofiltration (NF) and reverse

osmosis (RO), alongside Fenton and photo-Fenton processes (Shahid et al., 2021).

Interactions of pharmaceuticals with soil matrices depend on their properties such as molecular structure and spatial arrangement, hydrophobicity, polarity, polarizability (Zhi et al., 2019), polar surface area (PSA), as well as soil characteristics like soil type and pH, soil organic matter (SOM), and coexisting ions (Silva et al., 2021). Polar pharmaceutical sorbates mostly interact with SOM through H-bonds and van der Waals forces, while hydrophobic PPs do so through hydrophobic interactions, such as  $\pi$ - $\pi$  bonds and aromatic ring alignment (Ahmed et al., 2015). Applying the experimental log Kow values for three sulfonamides at pH 5 (for sulfadiazine: 0.14, sulfamethazine: 0.27, and sulfachloropyridazine: 0.69) reported by Carda-Broch and Berthod (2004), Conde-Cid et al. (2020) illustrated that hydrophobicity can better predict the transportation behavior of sulfonamide antibiotics compared to pKa/ionic species. Movement and transportation pathways within a more homogeneous group of PPs may not be applicable across different classes of PPs, and other characteristics like functional groups of the compounds can significantly influence their environmental persistence and mobility. The pronounced polarity and presence of multi-ionic groups are primarily responsible for the significant soil adsorption capacities exhibited by fluoroquinolones and tetracyclines (Zhi et al., 2019). On the other hand, the PSA can serve as a critical metric for evaluating the capacity of PPs to permeate cellular membranes. Silva et al. (2021) state that compounds exhibiting a PSA greater than 140 square angstroms (Å<sup>2</sup>) demonstrate diminished permeability across cellular barriers. Erythromycin, which has a PSA of 194 Å<sup>2</sup>, exemplifies this reduced permeability.

Cationic pharmaceutical species exhibit greater retardation compared to their acidic and neutral counterparts due to the predominantly negatively charged nature of soil surfaces (Schaffer et al., 2012). Various soil parameters significantly influence the adsorption capacities of PPs within soil environments; these include soil texture, clay content, cation exchange capacity (CEC), and anion exchange capacity (AEC). Several soil parameters influence the adsorption capacities of PPs to the soil, including soil texture, clay content and composition, and CEC or AEC. For example, due to greater surface area and CEC, enrofloxacin showed stronger adsorption to montmorillonite (expanded 2:1 clay minerals) than to kaolinite (1:1 clay minerals). The observed reduction in pharmaceuticals sorption with increasing pH can be elucidated through two mechanisms: alterations in charge dynamics (Srinivasan et al., 2013), and modifications in the sorption mechanism (Klement et al., 2018). In the former, as pH rises, sulfamethoxazole (SMX) loses some of its positive charges, and electrostatic repulsion reduces the sorption action; in the latter, in acidic soils, cation bridging and surface complexation of irbesartan (IRB) take the place of the CEC-driven mechanism. As a result, the repulsion of negative charges on the compounds and the soil surface greatly decreased sorption (Xu et al., 2021b).

Numerous investigations demonstrated the toxicity and bioaccumulation potential of certain PPs (Carter et al., 2016; Pullagurala et al., 2018; Almeida and Nunes, 2019). Antibiotics can harm cells, the circulatory system, the metabolism, and the development of fish, they have shown the ability to bioaccumulate in fish liver, lungs, and fillet and they have been detected in dairy products and milk samples and crops can absorb them (Bansal, 2022). Different fish species across various trophic levels selectively absorbed antidepressants. The most significant bioaccumulation occurred in the brain and was directly correlated with WWT effluent exposure. Following the brain, bioaccumulation was observed in the gonads, liver, and muscle tissues (Arnnok et al., 2017). Pérez et al. (2023) upheld pharmaceutical load as a useful bioaccumulation metric in the tissues of bulrushes (*Typha latifolia*). They showed that fluoxetine (FLX) accumulated more in the stem, carbamazepine (CBZ) and FLX in the leaf, and GMF in the root. The pharmaceutical mass load was related to log K<sub>ow</sub> in the root and rhizome, while in the leaf, it was correlated with pK<sub>a</sub> and plant transpiration.

## 4 Personal care products

Two categories of PCPs and their degradation byproducts were reviewed: parabens and chlorophenolic antimicrobials.

## 4.1 Overview

#### 4.1.1 Parabens

Parabens, or para-hydroxybenzoic acid esters, are synthetic compounds used as preservatives in cosmetics, pharmaceuticals, and food items since the 1920s. These substances effectively inhibit microbial proliferation and prolong shelf life (Ma et al., 2018; Chen et al., 2021; Stoiber, 2019). Their antimicrobial efficacy is particularly pronounced against molds, yeasts (Ye et al., 2006), and Grampositive bacteria (Stoiber, 2019). Parabens are EDCs with steroidogenesis disturbance and estrogenic effects (Liang et al., 2023) that can permeate the human body through dermal exposure when employing various products such as moisturizers, facial and body cleansers, UV sunscreens, deodorants, shaving creams, toothpaste, makeup, and other items that contain these substances (Stoiber, 2019), although environmental exposure has also been documented (Chen et al., 2021).

#### 4.1.2 Chlorophenolic antimicrobials

Like parabens, chlorophenolic antimicrobials triclosan (TCS), also known as irgasan, and triclocarban (TCC) have garnered significant attention from regulatory authorities due to their potential as EDCs (Chen et al., 2008) and for their contribution to the spread of multidrug resistance, including antibiotic (Carey and McNamara, 2014; Carey et al., 2016). For this reason, a phaseout notice of TCS and TCC from consumer antiseptic washes in the United States, and the ban on product-type 1 items (human hygiene biocidal products for skin or scalps) in Europe were issued in 2016 by the United States Food and Drug Administration, and the European Commission, respectively (USFDA, 2016; EC, 2016). The Food and Drugs Act in Canada authorizes TCS use in PPs at concentrations ranging from 0.1% to 1.0%. To safeguard human health, TCS is permitted at a concentration not exceeding 0.03% in mouthwashes, and at or below 0.3% in topical formulations and dentifrices (Health Canada, 2024). In October 2020, the Canadian

federal government issued a directive mandating that entities utilizing or importing 100 kg or more of TCS annually must develop a pollution prevention plan specifically for TCS. Individuals subject to the Final Notice in 2020 and 2021 are required to achieve 30% and 95% reductions, respectively, in the total mass of TCS utilized compared to levels established in the base year (Environment Canada, 2020). Regarding TCC, in light of the assessment that the risk it poses to human health is low, the government is contemplating the establishment of a monitoring plan for surface water and sediment, as part of the chemical management plan (CMP) (Environment Canada and Health Canada, 2023).

# 4.2 Physical and chemical characteristics: indices of fate and transport

#### 4.2.1 Parabens

Parabens are characterized by a benzoic acid backbone featuring a hydroxyl group positioned at the para-position. This structural configuration arises from the esterification of the carboxylic acid function with diverse alcohols (Soni et al., 2005; Ma et al., 2018), thereby contributing to their hydrophobic properties, which in turn influence their solubility (Ma et al., 2018; Chen et al., 2021; Li et al., 2020). The presence of hydroxyl group facilitates H-bonding, thereby enhancing their interactions with biological molecules and potentially contributing to their endocrine-disrupting effects. The significance of parabens as CECs lies in their ability to act as EDCs (Ma et al., 2018). Their ester bonds are attributed to their chemical stability as they can withstand hydrolysis in some situations, although they may break down in extremely acidic or alkaline conditions (Chen et al., 2021). Each paraben exhibits variability in the length and branching of its alkyl chains, which significantly affects its physical and chemical behavior (Chen et al., 2021). Generally, parabens display moderate solubility in water, a property that is contingent upon the length of the alkyl chain; short-chain parabens tend to exhibit greater solubility compared to their long-chain counterparts. This solubility characteristic has implications for their distribution and persistence within aquatic ecosystems and wastewater treatment systems. Additionally, these compounds possess relatively low molecular weights, which can influence their volatility and environmental mobility (Ma et al., 2018). The melting and boiling points of parabens are determined by their molecular structure and chain length, typically increasing with longer alkyl chains. While parabens are subject to degradation in environment, their persistence and behavior can be affected by various factors including temperature, pH levels, and microbial activity (Li et al., 2020).

#### 4.2.2 Chlorophenolic antimicrobials

TCS and TCC are chlorinated aromatic compounds characterized by monochlorophenyl and dichlorophenyl moieties. Both compounds exhibit low solubility in aqueous environments and possess high log  $K_{ow}$  values, with TCS at 4.8 and TCC at 4.9 (Coogan et al., 2007; Kwon and Xia, 2012). These properties contribute to their pronounced lipophilicity while reducing their hydrophilicity, indicating a pronounced inclination for partitioning into organic fractions rather than remaining in the aqueous phase (Carey and McNamara, 2014; Armstrong et al., 2019).

### 4.3 Occurrence in sewage sludge and soils

The occurrence of twenty PCPs grouped into preservatives (eight parabens and four degradation products) and antimicrobials (TCS, TCC, and six metabolites) was evaluated in sewage sludge and soils.

### 4.3.1 Parabens

The presence of six widely used parabens and 4 byproducts in WWT process and sewage sludge was examined by Ma et al. (2018). The analyzed paraben compounds are methylparaben (MeP), ethylparaben (EtP), propylparaben (PrP), butylparaben (BuP), benzyl paraben (BzP), and heptyl paraben (HepP), and the four metabolites are 4-hydroxybenzoic acid (4-HB), 3.4dihydroxybenzoic acid (3,4-DHB), methyl protocatechuate (OH-MeP) and ethyl protocatechuate (OH-EtP). Parabens have been detected in wastewater and sludge, indicating frequent contamination. Chen et al. (2021) investigated eight parabens -MeP, EtP, PrP, BuP, BzP, HpP, isopropyl paraben (isoPrP), and isobutyl paraben (isoBuP), and identified their presence in various outdoor environmental media, including soil samples. The prevalence of parabens is predominantly characterized by isobutyl-paraben (iso-BuP), succeeded by 4-hydroxybenzoic acid (4-HB) and 3,4-dihydroxybenzoic acid (3,4-DHB), with respective concentrations measured at 131 ng/g, 100 ng/g, and 86 ng/g d. w.

#### 4.3.2 Chlorophenolic antimicrobials

TCS and TCC were investigated by numerous authors in sewage sludge (Moško et al., 2021; Li et al., 2021; Chen et al., 2019; Malvar et al., 2020b; Malvar et al., 2020a) and soil (Mercl et al., 2021; Malvar et al., 2020b; Chen et al., 2021; Abril et al., 2018). TCC and TCS are the most prevalent PCPs in sewage sludge, with a median concentration of 1,710 and 1,165 ng/g, respectively. Additionally, degradation byproducts such as monocarbanilide (MCC), 2'-hydroxytriclocarban (2-OH-TCC), carbanilide (CBN), and 3,3',4,4'tetrachlorocarbanilide (TCCC) exhibited median concentrations of 520 ng/g, 180 ng/g, 102 ng/g, and 91 ng/g, respectively. These concentrations are within the quantifiable range of their parent compounds, emphasizing the necessity to investigate both the parent substances and their metabolites, as highlighted by Chen et al. (2019). The concentration ranges for examined parabens and antimicrobials in sewage are observed to be between 380 and 81,750 ng/g; notably, antimicrobials predominate over parabens, with the concentration range for the former extending from 132 to 79,985 ng/g. TCS and TCC in soil were below the quantitation limits in the researched studies. Statistics on the individual and cumulative occurrence of PCPs in sewage sludge and soil can be found in Table S2 of Supplementary Appendices A, B- Sheet 4-PCPs.

# 4.4 Environmental fate and transport mechanisms

#### 4.4.1 Parabens

The main ways that parabens enter the environment are through wastewater and runoff from homes where these compounds are utilized in various domestic items. In the WWT process, parabens can be successfully eliminated by the cyclic-activated sludge

technology and anaerobic-oxic (A/O) treatment methods (Ma et al., 2018). It has also been demonstrated that temperature and pH impact the biodegradation of parabens using aerobicactivated sludge. Indeed, parabens maintain stability in neutral to slightly acidic pH ranges; however, they are susceptible to degradation under extreme pH conditions, which is relevant in WWT processes. Increased residual quantities of parabens were reported at pH 10.0 compared to neutral or slightly acidic conditions, indicating decreased degradation efficiency (Ma et al., 2018). Higher temperatures cause the aqueous phase's residual concentrations to drop. Possibility of aerobic-activated sludge biodegrading parabens, emphasizing the role of this process in WWT (Lu et al., 2018). Parabens are readily biodegradable in soil and unlikely to threaten earthworms' survival and reproduction (Arachchige Chamila Samarasinghe et al., 2021). Nonetheless, due to their widespread use and pseudo-persistence, they are present in the environment and may be more dangerous for other species or toxicological endpoints. Parabens exhibit varied environmental fate as influenced by biodegradation, photodegradation, and sorption to particulate matter (Chen et al., 2021), and organic matter (Mercl et al., 2021). Their transport mechanisms primarily involve water pathways, volatilization into the atmosphere (Chen et al., 2021; Li et al., 2020) with potential contributions from atmospheric and soil movement. Some parabens may bioaccumulate, leading to higher concentrations in the tissues of biological organisms.

#### 4.4.2 Chlorophenolic antimicrobials

The characteristics of TCS and TCC significantly influence their removal efficiency during WWT processes. Kwon and Xia (2012) indicate that, owing to their high log  $K_{\mbox{\tiny ow}}$  up to 90% of TCS and TCC entering WWTPs are adsorbed onto the sludge during the treatment processes. In a simulation study, it was shown that variations in hydraulic retention time, sludge retention time and temperature influence the removal (SRT). and transformation of TCS and TCC throughout WWT (Armstrong et al., 2018). Apparently, both antimicrobials tend to adsorb onto sludge, resulting in minimal concentrations remaining in the effluent. Furthermore, their degradation rates increased with elevated temperatures, with TCS exhibiting a more rapid degradation than TCC. To learn more about the destiny of these two antimicrobials, a second WWT simulation was performed under nitrifying conditions. The findings indicated a rapid degradation of TCS into methyl triclosan (MeTCS), which is becoming increasingly pronounced in alkaline environments (Armstrong et al., 2019). Conversely, TCC exhibited significant resistance to this treatment. The emergence of MeTCS, a degradation product of TCS with a log Kow value of 5.2 (Coogan et al., 2007), suggests heightened concern due to its greater potential for bioaccumulation compared to the parent compound. Alongside WWT transformation products like carbanilides, MeTCS is thought to retain endocrine disruption capabilities (Ahn et al., 2008), raising additional ecological concerns.

Li et al. (2020) discussed the stability of TCS under typical environmental conditions but highlighted the possibility of photodegradation when exposed to ultraviolet light. In contrast, TCC is acknowledged for its lasting presence in the environment; it demonstrates resistance to degradation processes and has the capacity to accumulate in aquatic and terrestrial ecosystems following release via WWT operations (Carey and McNamara, 2014; Armstrong et al., 2019; Abril et al., 2018). As specified above, the presence of TCS and TCC was below the quantitation limits. This phenomenon may be attributed to their degradation potential, as postulated by Lozano et al. (2010), who observed a substantial reduction in TCS levels in agricultural soils 16 months following the application of biosolids. In a similar context, Anand et al. (2022) highlighted the biotransformation of parent CECs, as opposed to their elimination or biodegradation, as a plausible mechanism for elevating the concentration of total contaminants in effluent relative to the influent. Degradation byproducts of antimicrobials were not measured in the consulted studies. Lastly, the presence of parabens, TCS, and TCC has been determined in urine (Shin et al., 2019), human serum (Li et al., 2020; Assens et al., 2019), and in adipose tissue (Artacho-Cordón et al., 2018), highlighting their bioaccumulation potential. Pérez et al. (2023) demonstrated an elevated bioaccumulation of TCS in bulrush (T. latifolia) roots. In a separate study by Cavanagh et al. (2018), TCS demonstrated considerable potential for endocrine disruption, notably through its binding affinity to the transthyretin (TTR) receptor. It was also highlighted for its anti-androgenic properties, suggesting a capacity to interfere with normal developmental processes and male reproductive health.

## 5 Hormones

### 5.1 Overview

Natural and synthetic hormones are increasingly recognized as CECs due to their potential impacts on environmental and human health. Natural hormones, such as estrogens and androgens, are excreted by humans and animals and can enter water systems through wastewater (Adeel et al., 2017; Naldi et al., 2016). Additionally, synthetic hormones, such as those utilized in contraceptives, hormone replacement therapies (HRT) (Viglino et al., 2008; Koubovec et al., 2005), anabolic agents for muscle growth and strength enhancement (Liu and Wu, 2019; Bond et al., 2022), cancer therapy (Johnston and Cheung, 2018; Sartor and Gillessen, 2014), and various agricultural applications (Xuan et al., 2008), further contribute to environmental contamination. Hormones are biologically active at very low concentrations and can disrupt endocrine systems in wildlife, leading to reproductive and developmental abnormalities (Diamanti-Kandarakis et al., 2009). For example, exposure to estrogen hormones in aquatic environments has been linked to feminization of male fish (Jobling et al., 2006). Thus, hormones are a growing focus of environmental monitoring and regulatory efforts to mitigate their ecological and health impacts.

# 5.2 Physical and chemical characteristics: indices of fate and transport

Based on their chemical structure, hormones are threefold: lipidderived hormones (or lipid-soluble hormones), amino acid-derived hormones, and peptide hormones. Most lipid-soluble hormones are

derived from cholesterol and exhibit structural similarities. Steroid hormones are the main class of lipid hormones in humans. Steroid hormones estradiol (estrogen) and testosterone (androgen), which are associated with female and male sexual traits, respectively, are in this class. Chemically, they are categorized as ketones or alcohols. The amino acid-derived hormones are made of tryptophan and tyrosine amino acids, which are comparatively small molecules. Hormones in this group include thyroxine, which is produced by the thyroid gland, and adrenal gland medulla-made norepinephrine and epinephrine. In contrast, peptide hormones are characterized by their polypeptide chain structure, which consists of amino acids. They include oxytocin and antidiuretic hormones, which are synthesized in the brain and released into the bloodstream by the posterior pituitary gland. This category also encompasses smaller proteins, such as growth hormones produced by the pituitary gland, as well as larger glycoproteins like follicle-stimulating hormones (Fowler et al., 2018).

# 5.3 Occurrence in sewage sludge, biosolids, and soils

Research has demonstrated the occurrence of hormones in various environmental matrices, including soil (Kumirska et al., 2019), wastewater (Gewurtz et al., 2022) and biosolids (Lorenzen et al., 2004). Here, we analyzed studies focusing on hormonal compounds present in sewage sludge (Silva et al., 2021; Riva et al., 2021; Moško et al., 2021; Svahn and Björklund, 2019), biosolids (Gewurtz et al., 2022), and soil (Kumirska et al., 2019). The hormones under investigation include eleven naturally occurring hormones: androstenedione (A4), androsterone (AN), cortisone (E), estrone (E1), 17 β-estradiol (E2), 17 α-estradiol (17 a-E2), estriol (E3), progesterone (P), testosterone (T), equilin (EQL), and equilenin (EQN); as well as ten synthetic hormones: 17 aethinylestradiol (EE2), allyl trenbolone or altrenogest (ALT), desogestrel (DSG), diethylstilbestrol (DES), gestodene (GST), melengestrol acetate (MGA), mestranol (EEME), norethindrone (NRT), norgestrel (NRG), 17 a-dihydroequilin (2H-EQL). The quantification of four hormones in sewage sludge reveals a predominance of GST, with a concentration of 41 ng/g, followed by E1 at 17 ng/g, and E2 at 16 ng/g. In biosolids, the hormonal composition is primarily characterized by synthetic hormones, specifically DSG at 252 ng/g and EEME at 176 ng/g, while natural hormones are represented by E3 at 95 ng/g, AN at 54 ng/g, and P at 28.5 ng/g. Out of five hormones investigated in soils by Kumirska et al. (2019), E2, EE2, and DES appeared below the quantitation limits, while E1 and E3 showed respective median concentrations of 6.3 ng/g and 1.9 ng/g. Supplementary Table S3 gives detailed information on the occurrence of hormonal compounds in examined matrices.

# 5.4 Environmental fate and transport mechanisms

According to Limpiyakorn et al. (2009), peptide and steroid hormones represent significant environmental concerns. Hanselman et al. (2003) assert that endogenous hormones may exhibit extraordinary potency in disrupting the endocrine systems of organisms, potentially exceeding the effects of exogenous hormones by a factor ranging from 10,000 to 100,000. Various factors, including soil characteristics, organic matter content, pH levels, and microbial activity, can influence the sorption capacity of hormones to solid particles within WWT systems, thereby affecting their transport dynamics and potential removal (Ying and Kookana, 2005). The fate of these hormones is additionally modulated by residence time and temperature within WWT systems (Viglino et al., 2008). Numerous hormones, including both naturally occurring and synthetic variants, are not entirely eliminated during conventional WWT processes (Bai and Acharya, 2019), permitting their entry into adjacent water bodies and groundwater sources (Zhou et al., 2012). To cite an example, Huang et al. (2014) evaluated the removal efficacy of one progestogen (P), three androgens (A4, T, and dihydrotestosterone (DHT)), and four estrogens (E1, E2, E3, and EE2) throughout the WWT process. Their findings indicated a remarkable removal efficiency for androgens and progestogens, ranging from 86% to 100%. In contrast, the removal efficiency observed for estrogens was comparatively lower, falling within the 75%-92% range.

It is established that natural hormones are excreted by living organisms in conjugated forms that remain undetectable by analytical methods designed for unbound hormones. Given that conjugation can be reversed by naturally occurring enzymes present in WWTPs and the environment, the ecological fate and migration mechanisms of these hormones are notably affected by both conjugation/deconjugation processes as well as the specific wastewater treatment methodologies employed (Gewurtz et al., 2022). Deconjugation of some of the hormones excreted in urine seems to occur within a time frame of hours to days, within the period when the WWT process may have a significant impact on deconjugation (Naldi et al., 2016). Hormones demonstrate persistence against various treatment approaches (e.g., sedimentation, biological processing, and disinfection), leading to their detection in effluents released into ecosystems. This situation raises concerns over EDCs like E2, which can pose substantial ecological risks, including fish feminization and disruption of aquatic ecosystems, even at concentrations as minimal as 2 ng/L (Bai and Acharya, 2019). However, when wastewater undergoes treatment through techniques such as ultrafiltration (UF) or ozonation, research has indicated varying degrees of efficacy regarding hormone removal. For instance, the algae-mediated degradation exhibited a notable removal efficiency of approximately 60% for certain hormones like E2 and EE2. Gewurtz's study (2022) explained that WWT techniques such as lagoon systems alongside secondary and advanced treatment facilities could initiate hormonal deconjugation processes impacting concentrations of hormones like EEME and T. Furthermore, this investigation highlights that upon discharge into aquatic environments or application of treated biosolids on agricultural lands, these conjugated hormonal forms may subsequently undergo deconjugation in the natural environment. Such processes could enhance hormonal bioavailability, allowing for increased interactions with aquatic ecosystems and soils, which might affect terrestrial flora and fauna uptake.

The transport dynamics of hormones within soil matrices are contingent upon their solubility profiles, adsorption affinities to soil

particles, as well as uptake mechanisms involving plants or microorganisms (Bai and Acharya, 2019). The deconjugated form of these hormones exhibits lipophilicity, whereas their conjugated counterparts are hydrophilic (Gewurtz et al., 2022); this distinction illustrates how hormonal compounds may leach into groundwater or adhere to soil particles. Hormones may be either adsorbed onto particulate matter or dissolved within soil solution. Extended retention times in soils can allow for the degradation of certain compounds. Still, retention on soil surfaces would reduce the solution concentration of hormones and slow down their degradation. Furthermore, hormones may also migrate through leaching or surface runoff pathways, leading to potential groundwater contamination or infiltration into surface water bodies. In environmental contexts, plant systems can absorb hormones, potentially influencing physiological functions such as root development, shoot growth patterns, flowering events, and germination processes (Shi et al., 2010). Moreover, hormonal compounds have been documented to bioaccumulate in species such as quagga mussels (Dreissena bugensis), indicating their propensity to infiltrate food webs (Bai and Acharya, 2019), a phenomenon posing considerable risks to ecological health. Microbial transformations also play a crucial role in facilitating hormone degradation or transformation, including analogous deconjugation phenomena observed within WWTPs (Gewurtz et al., 2022; Olsen et al., 2007). Czajka and Londry (2006) highlighted the susceptibility of hormones to microbial degradation within soil environments depending on specific bacterial populations and other variables such as moisture levels and soil composition types; they further documented hormones' anaerobic transformation capabilities under certain conditions within soil matrices. Bartelt-Hunt et al. (2012) emphasized concerns associated with runoff occurrences in regions characterized by intensive animal husbandry practices where manure applications could directly introduce hormones into local ecosystems.

## 6 Plastic-related compounds

Bisphenols (BPs) and phthalic acid esters (PAEs) are two classes of plastic-related compounds under scrutiny. The literature examined documented the presence of twenty BP analogs and fifteen PAEs within the soil and sewage sludge matrices.

## 6.1 Overview

#### 6.1.1 Bisphenols

Phenolic plastic-related compounds, such as bisphenol A (BPA), are synthetic compounds employed to enhance the flexibility and durability of plastics (Kubwabo et al., 2009; NIEHS, 2023). These substances are common in polycarbonate plastics and epoxy resins, utilized across various consumer products, including water bottles, food packaging, and medical devices (EFSA, 2023; Park et al., 2018). Research has raised concerns about the potential health repercussions of bisphenols, particularly BPA. This compound has been implicated in endocrine disruption, reproductive disorders, and an elevated risk of certain cancers (vom Saal et al.,

2007; Lang et al., 2008). Furthermore, studies have demonstrated that BPA can leach from plastic materials into consumables, thereby facilitating human exposure (FDA, 2023). In light of these apprehensions, several states have instituted bans on using BPA in specific products such as baby bottles and sippy cups (EC, 2011a; EC, 2011b). Concurrently, various manufacturers have proactively substituted BPA with alternative bisphenols like bisphenol S (BPS) or bisphenol F (BPF). Nevertheless, research has also raised alarms about the potential health implications associated with these alternative bisphenols. A recent study correlated obesity in young people to exposure to BPS and BPF (Jacobson et al., 2019). Another study examined the endocrine disruption potential of six commercially available BPA alternatives: BPS, BPF, bisphenol AP (BPAP), bisphenol AF (BPAF), bisphenol Z (BPZ), and bisphenol B (BPB). All 6 BPA substitutes were found to be no less estrogenic than BPA in human breast cancer cells, with BPAF, BPB, and BPZ being even more estrogenic (Mesnage et al., 2017). As a consequence, experts are advocating for minimizing exposure to all bisphenols while promoting safer alternatives (Rochester and Bolden, 2015; Viñas and Watson, 2013).

#### 6.1.2 Phthalic acid esters

PAEs, called phthalates, are synthetic compounds widely employed as plasticizers across various consumer products. These include flexible polyvinyl chloride (PVC) items such as vinyl flooring, wallpaper, and apparel; personal care items encompassing cosmetics, fragrances, and lotions; medical apparatus including tubing, catheters, and gloves; as well as food packaging and processing equipment (Wang and Qian, 2021; Lee et al., 2019; Net et al., 2015; Li et al., 2017). The mechanism by which phthalates function involves enhancing the flexibility and durability of plastics, thereby rendering them more malleable and resistant to fracturing. Nonetheless, these substances are not chemically integrated within the plastic, which renders them susceptible to leaching into the environment and potentially into foodstuffs and human organisms via ingestion, inhalation, or dermal contact matrix (Wang et al., 2022). Such leaching may result in human exposure that raises significant health concerns attributed to their potential endocrine-disrupting characteristics, which can interfere with human hormonal systems (Zhang et al., 2021a; Zhou et al., 2019). Empirical studies have established associations between phthalate exposure and adverse reproductive outcomes, congenital anomalies, as well as developmental disorders (Swan Shanna et al., 2005). For this reason, regulatory entities such as the European Chemicals Agency (ECHA) and the USEPA have prohibited or subjected PAEs to restrictions (ECHA, 2022; USEPA, 2023a).

# 6.2 Physical and chemical characteristics: indices of fate and transport

#### 6.2.1 Bisphenols

The chemical structure of BPs consists of two phenolic groups connected by a bridging group (such as carbon or sulfur) (Sánchez-Piñero et al., 2020; Xu et al., 2021a). This structure contributes to their chemical reactivity and ability to form polymers, which accounts for their extensive application as plastic-related compounds (Xu et al., 2021a). BPs tend to be soluble in organic solvents but exhibit limited solubility in water (Zhu et al., 2019; Sun et al., 2018; Huang et al., 2020). This hydrophobic behavior affects their distribution in sludge and increases their potential for adsorption onto particulate matter (Zhu et al., 2019).

#### 6.2.2 Phthalic acid esters

PAEs constitute a group of industrial compounds distinguished by their chemical composition comprising two alkyl or aryl groups bonded to a phthalate group. Their unique physicochemical characteristics render them suitable for diverse applications. Predominantly employed as plasticizers or additives in consumer goods, PAEs can be classified based on their molecular weight. Higher molecular weight phthalates are typically utilized as plastic additives to enhance the pliability and durability of polyvinyl chloride (PVC) products, exemplified by DEHP. Lower molecular weight phthalates fulfill various functions (e.g., solvents, emulsifiers, stabilizers) in consumer items like cosmetics, personal care products (PCPs), insecticides, adhesives, pharmaceuticals, and solvents - examples being diethyl phthalate (DEP), di-n-butyl phthalate (DnBP), and dimethyl phthalate (DMP) (Koniecki et al., 2011; Tao et al., 2023).

### 6.3 Occurrence in sewage sludge, and soils

#### 6.3.1 Bisphenols

Publications on twenty BPs in sewage sludge and the soil were examined. BPA and BPF are the most dominant sewage sludge among BP analogs, with 179 ng/g and 165 ng/g median concentrations, respectively. BPF (3.4 ng/g) and BPA (2.3 ng/g) dominate again in the soil. The median total concentration of twenty bisphenol analogs in sewage is 388 ng/g, and 9.6 ng/g in the soil. Detailed data are found in Supplementary Table S4.

#### 6.3.2 Phthalic acid esters

Fifteen PAEs were analyzed in sewage sludge or soil by several authors. PAEs represent the most dominant class of CECs occurring in sewage sludge and soil compared to all other classes reviewed in this paper. In fact, the median cumulative concentration of 15 PAEs in sewage sludge is 734  $\mu$ g/g, with minimum and maximum values ranging from 107  $\mu$ g/g to 2,618  $\mu$ g/g. The concentrations of PAEs are consistently elevated in sewage sludge when compared to those found in soils. The median concentration of PAEs measured in the soil is 2.99  $\mu$ g/g, with the lower and upper optima of 0.28  $\mu$ g/g and 634  $\mu$ g/g, respectively. DEHP registered the highest median concentration in sewage (224  $\mu$ g/g), followed by BBzP (202  $\mu$ g/g), and dibutyl phthalate (DBP) (182  $\mu$ g/g). In the soil, DEHP dominates (1.73  $\mu$ g/g), followed by DiBP (0.53  $\mu$ g/g), and DBP (0.29  $\mu$ g/g). Additional information is in Table S6 and Supplementary Appendix B-Sheet 7-Phthalates.

# 6.4 Environmental fate and transport mechanisms

#### 6.4.1 Bisphenols

Understanding the environmental behaviors and mobility of BPs is essential for assessing their environmental impact and developing strategies to mitigate their adverse effects. The fate of BPs in the WWT process, sewage sludge, and soil is influenced by a complex interplay of factors, and it varies depending on various factors such as their physicochemical properties, environmental conditions, and treatment methods. Studies have shown that BPs are not completely removed during conventional WWT, leading to their presence in both sewage sludge and liquid effluents (Huang et al., 2020). BPs may partition into the water phase, sorb onto solid fractions during WWT, and accumulate in sewage sludge (Xu et al., 2021a; Peng et al., 2020). During WWT and in soil, BPs undergo various transformations, influencing their environmental fate and transportation mechanisms (Sun et al., 2018; Zhu et al., 2019; Sánchez-Piñero et al., 2020). Xue and Kannan (2019) have noted that BPs are not completely removed during the activated sludge treatment process; a substantial portion becomes bound to solids within the sewage sludge during treatment, which subsequently facilitates their transportation to land when this sludge is utilized as fertilizer in agricultural practices (Zhu et al., 2019).

Considerable disparities exist in the removal efficiencies of BPs, especially BPA, across different WWT technologies. Wang et al. (2019) illustrate that primary treatment exhibits the lowest BPA removal rate, averaging approximately 25.4%, followed by lagoon treatment with a moderate removal efficiency averaging 58.3%. Biologically aerated filters demonstrate enhanced removal efficiency with an average rate of around 66.0%. Finally, the activated sludge process yields superior performance with an average removal rate of 74.7%. Additionally, the Oxidation Ditch (OD) process marginally outperforms modified activated sludge processes, such as Anaerobic-Anoxic-Oxic (AAO) and Anoxic-Oxic (AO) methods), which exhibit slightly lower removal efficiencies compared to standard activated sludge.

Upon application of sewage sludge as fertilizer or soil amendment, BPs can be released into the soil environment. Studies have identified BPs in sewage sludge (Peng et al., 2020; Sun et al., 2018; Zhu et al., 2019) and agricultural soils (Xu et al., 2021a; Sánchez-Piñero et al., 2020). The accumulation of BPs in soil from biosolids raises concerns about their persistence, estrogenic potency, and potential risks associated with land application. Several factors influence the fate of BPs in biosolids following their introduction into soil environments. Weaker binding may facilitate leaching into groundwater systems; conversely, strong adsorption can inhibit such occurrences. Exposure to environmental factors within soils can prompt further degradation of BPs through microbial action or photodegradation processes. Factors such as temperature, moisture content, and soil type can all influence degradation rates (Zhu et al., 2019; Sánchez-Piñero et al., 2020). Generally, BPs are chemically stable under certain conditions but may undergo hydrolysis or degradation under extreme pH, temperature, or oxidative conditions, impacting their persistence in the environment and potential biological effects (Zhu et al., 2019; Xu et al., 2021a). A study conducted in Scotland and the United Kingdom showed that 4% by mass of BPA in the soil was predicted to enter soil pore water, resulting in significant uptake by crops, primarily leafy vegetables, but also root crops, and much lower uptake into cereal grains (Zhang et al., 2015).

#### 6.4.2 Phthalic acid esters

U.S. EPA's list of priority pollutants outlines six PAEs including the two widely used ones, DBP and DEHP (Wang

et al., 2022; USEPA, 2023b). The primary sources of PAEs in soils comprise the application of biosolids, sewage irrigation, and chemical fertilizers and additives (Wei et al., 2020; Becky Miriyam et al., 2022). The transport pathways and environmental destiny of PAEs within soil and air matrices are influenced by a confluence of factors, including the origins of pollution, their respective properties, the inherent properties of the soil, and its associated microbiological characteristics (Tuan Tran et al., 2022; Koniecki et al., 2011; Wang et al., 2022). Certain PAEs, particularly those of lower molecular weight, possess heightened volatility (e.g., DEHP Henry's law volatility constant value of  $4.37 \times 10^{-5}$  atm-m<sup>3</sup>/mol) potentially resulting in their release into the atmosphere and subsequent exposure via inhalation or dermal contact (Koniecki et al., 2011; Howard, 2017).

Wang et al. (2022) studied the impact of soil-particle size on the migration of PAEs in soil medium. They found that total PAE concentrations in bulk soil decreased as samples were collected farther from point-source pollution, in this example, the landfill. Clay-sorbed PAEs (clay particle size  $< 2 \mu m$ ) were found to be consistent with the Gaussian air pollution model, suggesting atmospheric transportation. Silt-sorbed (2 µm < particle size < 63 µm) PAEs showed a leveled-off concentration, suggesting shorter transport distance due to gravitational deposition, explained by the Boltzmann equation. Sand-sorbed PAEs (particle size > 63  $\mu$ m) showed an unexpected increasing trend in surrounding soils, emphasizing the apparent accumulation of PAEs at downhill sites. When PAEs interact with trace elements in soils, their level of toxicity is raised (Zhang et al., 2021b). Berenstein et al. (2022) found that phthalic acid and monobutyl phthalate (MBP) are generated by the photodegradation of DBP in polyethylene mulches. The same research report that the migration of DBP and MBP from these plastic covers into the surrounding environment constitutes a notable concern. Another research by Zeng et al. (2008) pointed out that the PAEs' metabolites can exhibit toxicity levels similar to those of their parent compounds. It has been observed that there exists a positive correlation between the lipid content of vegetables and the bioconcentration factor (BCF) of PAEs (Wei et al., 2020). Furthermore, it is unlikely that complete mineralization of PAEs will occur under in situ conditions within biosolidsamended soils (Madsen et al., 1999). The airborne migration of PAEs significantly elevates human exposure levels. The dermal absorption rates are contingent upon the specific types of phthalates, with DEP demonstrating particularly high absorption potential. This underscores its likelihood of contributing to cutaneous exposure through cosmetic and personal care products. Extensive research has established a correlation between PAEs and various health concerns, especially regarding their capacity to disrupt endocrine functions as well as adversely affecting reproductive and developmental processes, and potentially pose carcinogenic risk (Wei et al., 2020; Koniecki et al., 2011; Kaewlaoyoong et al., 2018; Yoshida et al., 2020). Given their ubiquitous presence in the environment, PAEs raise critical concerns about chronic exposure within the general population, particularly among vulnerable demographics such as infants and toddlers (Koniecki et al., 2011).

## 7 Rubber and polymer antioxidants [Aromatic secondary amines (Ar-SAs)]

### 7.1 Overview

Two main categories of aromatic secondary amines (Ar-SAs) are commonly utilized as antioxidants in rubber and polymers: substituted diphenylamines (S-DPAs) and substituted p-phenylenediamines (S-PPDs) (Kassler et al., 2014; Lu et al., 2016; Liu et al., 2019).

#### 7.1.1 Substituted diphenylamines

S-DPAs form a group of secondary amine antioxidants that are widely employed in petroleum-based products (fuels, lubricants), rubber materials, plastics, and other polymeric substances to prevent oxidative deterioration (Kassler et al., 2014). The diphenylamine (DPA) central structure is the basis and the lone functional group all S-DPAs share. The DPA's phenyl rings can have one to four side substitutions that are either phenyl or saturated alkyl, with the alkyl side chains containing four to nine carbons (OECD, 2016). In addition to the PREPOD (2-Propanone, reaction products with diphenylamine, CAS RN 68412-48-6) group investigated by Zhang et al. (2020c), three other remarkable classes of S-DPA antioxidants can be recognized as alkylated (mono-, di-, or poly-) DPAs [e.g., BNT (Benzenamine, N-phenyl-, reaction product with 2,4,4-trimethylpentene), CAS RN 68411-46-1], S-DPAs of variable numbers of phenyl/benzyl replacements on DPA rings [e.g., BNS (Benzenamine, N-phenyl-, styrenated), CAS RN 68442-68-2], and S-DPAs with changing numbers of alkyl/benzyl and phenyl substitutions [e.g., BNST (Benzenamine, N-phenyl-, reaction products with styrene and 2,4,4 trimethylpentene), CAS RN 68921-45-9] (Health Canada, 2010). These classes are mainly UVCB (Unknown or Variable Composition, Complex Reaction Products, and Biological Materials) chemical substances.

#### 7.1.2 Substituted p-phenylenediamines

S-PPDs, on the other hand, are added to the rubber as antioxidants, anti-ozonolysis, and bending crack inhibitors (Huntink et al., 2004). Huang et al. (2021) mentions three forms of PPDs utilized in the rubber processing industry, which are the dialkylated PPD (N,N'-dialkyl-p-phenylenediamine), the monoalkylated-mono-arylated PPD (N-alkyl-N'-aryl-pphenylenediamine), and the diarylated PPD (N,N'-diaryl-pphenylenediamine). The UVCB class of BENPAT (1,4-Benzenediamine, N,N'-mixed phenyl and tolyl derivatives, CAS RN 68953-84-4) studied by Zhang et al. (2020c) and captured on Canada's List of all Challenge chemical substances (Health Canada, 2010), alongside BENTAX (1,4-Benzenediamine, N,N'-mixed tolyl and xylyl derivatives, CAS RN 68478-45-5) exemplify S-PPDs.

### 7.2 Physical and chemical characteristics

#### 7.2.1 Substituted diphenylamines

According to an evaluation report of S-DPAs released by the Canadian federal government in 2017, empirical and modeled determinations of the S-DPAs' physicochemical characteristics displayed high log K<sub>ow</sub> values and poor water solubility (ECCC,

2017). For different S-DPAs, the anticipated log Kow values range from 4.45 to 13.58; however, the experimental measurement of log Kow values beyond 8.2 was uncertain because it is difficult to determine the partitioning accurately properties of superhydrophobic substances. The conjugate acid's acid dissociation constant (pK<sub>a</sub>H) of S-DPAs that are of 0.8  $\pm$ 0.4 indicates that, in an aqueous environment, the neutral form will predominate (ECCC, 2017). Overall, the physicochemical attributes and kinetics of S-DPAs are reported to be influenced by the degree of substitution and the number of carbon atoms in the side chains (OECD, 2016). S-DPAs typically have higher molecular weights than S-PPDs, affecting their volatility and solubility (Liu et al., 2019).

#### 7.2.2 Substituted p-phenylenediamines

The characteristics of S-PPDs, particularly in their roles as antioxidants and antiozonants, exhibit considerable variability based on the substitutions present on the nitrogen atoms. These diverse substitutions significantly impact their solubility, volatility, and reactivity profiles. S-PPDs demonstrate a range of solubility levels in aqueous environments. They are relatively stable under normal conditions but can undergo oxidation, especially when subjected to ozone or UV radiation, leading to the formation of secondary products like N-(1.3-dimethylbutyl)-N'-phenyl-pphenylenediamine (6PPD) (Huang et al., 2021). The polarity of these compounds is influenced by the presence of functional groups such as hydroxyl and amine moieties or other polar substituents that enhance their polarity, thereby affecting their behaviors in the environment (Moschet et al., 2018; Huang et al., 2021). The chemical structure of S-DPPs determines their stability. Bulky substituents on the aromatic ring can increase stability and decrease volatility. The essential component of these antioxidants' activity is their reactivity with oxidizing agents like peroxy radicals, produced when rubber ages. They can scavenge radicals, thus effectively terminating oxidative chain reactions (Huntink et al., 2004). The widespread use of these antioxidants in tire formulations and their resultant environmental discharge highlight their persistence and potential accumulation (Tian et al., 2021; Zhang et al., 2021c).

# 7.3 Occurrence in sewage sludge, biosolids, and soils

#### 7.3.1 Substituted diphenylamines

Fourteen S-DPAs, including six PREPODs and eight BNSTs, have been characterized in biosolids by Zhang et al. (2020c), Zhang et al. (2021c). The central tendency of the most occurring S-DPAs in biosolids is 86 and 66 ng/g for TO-DPA and DTOS DPA, respectively, while the central tendency for the least prevalent is 3.8 and 4.9 ng/g for IPDM-AD and IP-DPA, respectively. Overall, BNST compounds are the most recurring compared with the PREPODs, with a median concentration of 408 ng/g for the former and 43 ng/g for the latter. The maximum concentration of S-DPAs can achieve 3,133 ng/g of dry biosolids. Supplementary Table S8 gives information about the concentrations of S-DPAs quantified in indoor and playground dust. The most common among measured S-DPAs are di-n-octyl-DPA and 4,4'-bis(1,1-

dimethylbenzyl)diphenylamine (diAMS), with median concentrations of 74 and 46 ng/g in dust. The cumulative median concentration of the six measured S-DPAs was 215 ng/g, while the maximum concentration reached 13,619 ng/g. Supplementary Tables S6, S7 present data on the occurrence of S-DPAs in biosolids and dusts.

#### 7.3.2 Substituted p-phenylenediamines

This review investigates thirteen S-PPDs in biosolids or dust matrices, and respective occurrences are reported in Supplementary Tables S8, S9, respectively. N,N'-diphenyl-p-phenylenediamine (DPPD), N-phenyl-N'-(o-tolyl)-p-phenylenediamine (PTPD), and N,N'-di (o-tolyl)-p-phenylenediamine (DTPD) are three S-PPDs analyzed in biosolids totaling median concentration of 45 ng/g. Their concentration range varies from 4.5 to 366 ng/g. Twelve S-PPDs analyzed in dust showed a median concentration of 233 ng/g, with min-max spanning from 85 to 1,735 ng/g. The occurrence of S-PPDs in dust is dominated by N,N'-di (o-tolyl)p-phenylenediamine-quinone (DTPD-Q), N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine-quinone (6PPD-Q), and 6PPD, with respective concentrations of 60 ng/g, 42 ng/g, and 42 ng/g. The occurrence of DPPD in biosolids is to a certain extent similar as that found in dusts (15 ng/g in biosolids vs. 12 ng/g) and points out that there are several ways of exposure to this class of contaminants. Detailed statistical calculus is given in Supplementary Appendix B-Sheet 9-S-PPDs.

Research indicates that average concentrations of S-PPDs are significantly elevated in rubber playground dust in comparison to indoor dust, suggesting a heightened level of exposure in outdoor environments (Liu et al., 2019). Furthermore, another investigation revealed that S-PPDs, including 6PPD and its transformation product 6PPD-Q, are commonly found in dust samples collected from diverse settings, with the highest concentrations identified in road and vehicle dust (Huang et al., 2021). A study conducted in Tokyo, Japan, illustrated that the levels of 6PPD-Q were more pronounced in dust samples obtained from high-traffic areas than those from low-traffic regions, thereby demonstrating a significant influence of traffic patterns. Additionally, another study noted that concentrations of 6PPD-Q reached their zenith during months characterized by elevated atmospheric ozone levels (Hiki and Yamamoto, 2022).

# 7.4 Environmental fate and transport mechanisms

#### 7.4.1 Substituted diphenylamines

S-DPAs have been found in biosolids at amounts ranging from hundreds to over 1,000 ng/g dw in biosolid samples (Zhang et al., 2020c). In their studies, Zhang et al. (2020c), Zhang et al. (2021) successfully reported seventeen S-DPA in various compartments, including biosolids. The prevalence of S-DPAs in WWTP influent was positively correlated with the sizes of the served populations, indicating that the amount of waste produced significantly influences the concentration of S-DPAs in the treatment system (Lu et al., 2017). Sorption to sludge was identified as the primary mechanism for S-DPAs removal within WWTPs, with longer retention times in treatment units facilitating more efficient

partitioning of S-DPAs to solid sludge. Despite relatively high removal efficiencies, notable concentrations of S-DPAs were still detected in effluents (Zhang et al., 2020b; Lu et al., 2017). The biological treatment phases within the anoxic/oxic (A/O) process and cyclic activated sludge technology (CAST) have exhibited a higher efficiency in removing most S-DPAs than the hydrolysis acidification tank. This initial chemical-assisted treatment showed lower efficacy, likely due to biological processes enabling longer retention times, enhanced interaction between bacteria and pollutants, and the superior ability of microorganisms to degrade S-DPAs compared to purely chemical approaches (Zhang et al., 2020b). The research has also determined that meteorological elements, such as temperature and sun radiation, can influence the microbial populations' behavior and biodegradation mechanisms, consequently impacting the adsorption of S-DPAs on the sludge fraction.

Higher loads in benthic invertebrates (crayfish, Orcoescties spp.) than in pelagic fish (hornyhead chub, Nocomis biguttatus) and common shiners (Luxilus cornutus) indicate that the partitioning of S-DPAs in sediments was higher than in water (Lu et al., 2016). Likewise, concentrations of S-DPAs in biosolids surpassed those found in effluents, confirming that these compounds potentially accumulate and persist in the solid wastes generated during WWT (Zhang et al., 2016; Lu et al., 2017). This raises concerns about their long-term environmental impacts if these solids are used for land application or disposal. Given the substantial levels of S-DPAs persisting in biosolids, there are apprehensions regarding their beneficial reuse, particularly concerning practices like composting or land application. The presence of S-DPAs may result in their integration into terrestrial ecosystems and subsequent amplification throughout the food chain. This was evidenced by the detection of S-DPAs in the livers and eggs of seabirds, as well as in the livers of seals, indicating pollution within the Arctic ecosystem by this particular group of organic contaminants (Lu et al., 2019). Lastly, compared to seal livers, seabird livers, more particularly, the livers of the northern fulmar and black-legged kittiwakes, showed noticeably higher levels of S-DPAs, suggesting that these seabirds may be more vulnerable to exposure due to their migratory nature. Also, compared to seabirds, which have established migratory patterns, ringed seals showed greater individual variability in their migration patterns, which may affect the quantities and compositions of contaminants in their tissues (Lu et al., 2019).

#### 7.4.2 Substituted p-phenylenediamines

There is documented evidence that dust is an important sink for S-DPAs and S-PPDs in both indoor and outdoor settings (Hwang et al., 2016; Moschet et al., 2018), highlighting grime as a human exposure pathway through oral ingestion, skin exposure, and breathing (Mercier et al., 2011). S-PPD antioxidants are acknowledged as the most commonly encountered allergens among various rubber chemicals (Mahler, 2021). The recent revelation of the aquatic toxicity of 6PPD has been substantiated by its demonstrated harmful effects on fathead minnows (*Pimephales promelas*) (Prosser et al., 2017), along with the confirmed lethality of its oxidation byproducts, 6PPD-Q, towards coho salmon (*Oncorhynchus kisutch*) (Tian et al., 2021). Having said that, their environmental fate and transport mechanism are still unclear (Huang et al., 2021).

## 8 Pesticides

### 8.1 Overview

Here, literature pertaining to the presence of neonicotinoid insecticides (NEONICs) was reviewed. NEONICs are neuroactive and systemic insecticides that bear a structural resemblance to nicotine. Their widespread application in agriculture dates back to the early 1990s (Kollmeyer et al., 1999; Akter et al., 2023). NEONICs specifically target the sodium/ potassium ionophore within an insect's brain-spinal cord axis, disrupting cholinergic neural signaling by acting as agonists on nicotinic acetylcholine receptors (nAChRs). Upon binding tightly to these receptors in insects' central nervous apparatus, NEONICs can result in receptor blockage, paralysis, or fatality at higher concentrations while inducing neurological activation at lower levels. Notably, NEONICs exhibit a higher affinity for binding with insects' nAChRs than mammalians, rendering them preferentially more toxic to insect populations (Tomizawa and Casida, 2005). Agricultural practitioners and horticulturists highly favor NEONICs for several key reasons. They exhibit a wide-ranging efficacy against various insect pests, can be conveniently administered through various methods (seed treatments, soil drenches, or foliar applications), possess minimal acute toxicity towards mammals, and demonstrate systemic properties that enable plant uptake and distribution throughout plant tissues. This systemic nature provides safeguarding capabilities against pests that target different components of plants (Akter et al., 2023).

# 8.2 Physicochemical and biological characteristics

NEONICs exhibit a low n-octanol-water partition coefficient (log  $K_{ow} < 1$ ), an acid dissociation constant that lies considerably below or above the environmental pH ( $pK_a \le 2.2$  and  $pK_a \ge 11$ ), and a half-life ( $DT_{50}$ ) that spans from days to years (1–1,155 days). Supplementary Table S11 gives the log Kow, pKa, and half-life (DT50) values of NEONICs and the associated phenyl-pyrazole fipronil. Several authors have employed these physicochemical properties to explain the environmental fate and transportation mechanisms of NEONICs in soil and water media (Simon-Delso et al., 2015; Hladik and Kolpin, 2015). The N-nitroguanidines (clothianidin, dinotefuran, imidacloprid, and thiamethoxam), nitromethylenes and N-cyanoamidines (nitenpyram), (acetamiprid and thiacloprid) are the three chemical categories into which they can be placed (Jeschke et al., 2011).

# 8.3 Occurrence in sewage sludge, biosolids, and soils

Using a business espresso maker and a pressurized hot water extraction (PHWE) inox steel column, Svahn and Björklund (2019) examined the extraction of imidacloprid (IMI) alongside various pharmaceutical products (PPs) from biosolids. The findings indicated that the two extraction methods resulted in concentrations of 1.4 ng/g d. w. and 3.3 ng/g d. w. for IMI,

respectively. The presence of NEONICs was documented in various soil types and uses in China by Zhang et al. (2020a), Ying et al. (2022), Zhou et al. (2018), Zhou et al. (2021), and in Belize and the Philippines by Bonmatin et al. (2019), Bonmatin et al. (2021). Supplementary Table S12 summarizes statistics on quantified NOEs in the soil. A comprehensive review of eight NOEs in the soil compartment reveals key statistical measures. IMI demonstrates a higher median concentration of 2.6 ng/g, followed by acetamiprid (ACE) and nitenpyram (NIT) with 2.6 ng/g and 2.5 ng/g, respectively. Imidaclothiz (IMT) was found to be below the LOD. Summing up all NEONICs, the total ( $\Sigma_8$  NEONICs) exhibits a minmax interval of 0.372–273 ng/g, a median of 13 ng/g, and an average concentration measured at 25 ± 44 ng/g. Detailed statistics on quantified are found in Supplementary Appendix B-Sheet 10-Neonicotinoids.

# 8.4 Environmental fate and transport mechanisms

Because NEONICs dissolve in water, runoff and leaching are the usual ways through which they are transported. Plants may readily absorb them through their leaves or roots and then disperse them throughout their tissues (Borsuah et al., 2020; Pietrzak et al., 2020). Much of the applied NEONICs is lost to the surrounding soil, water, and air, with just about 5% being absorbed by the target plant (Wood and Goulson, 2017). In a leaching soil column study by Aseperi et al. (2020), the sorption potential of various NOEs in soil has been seen to differ. Specifically, thiacloprid (THA) exhibited soil adsorption up to 186 times higher than thiamethoxam (THI). Furthermore, it has been shown that the more organic matter in the soil, the more deeply NEONICs can accumulate in the soil after several applications and become persistent (Briceño et al., 2024).

## 9 Discussion

This review paper evaluated the presence, fate, and transportation mechanisms of various CECs grouped into pharmaceutical products (PPs), personal care products (PCPs), hormones, plastic-related compounds, rubber and polymer thorough antioxidants, and neonicotinoid insecticides. A methodology involving cross-referencing and thematic classification of existing literature provided a nuanced understanding of CECs in different environmental contexts. Figure 3 outlines the 50 most abundant CECs in biosolids, which are dominated by PAEs, followed by antifungals, chlorophenolic antimicrobials, fluoroquinolone antibiotics, and analgesics. Figure 4 provides a comprehensive overview of the occurrence of leading groups of CECs in sewage sludge or biosolids, with a predominant presence of PAEs, followed by PPs, PCPs, hormones, antioxidants, and bisphenols. Environmental transport and transformation processes are predominantly governed by factors such as solubility in water, volatility, biological and physicochemical degradation, sorption, and the potential for bioaccumulation, as described in Table 1. The findings aim to inform regulatory frameworks, illustrating how the cumulative presence of various groups of CECs in sewage sludge and soil could influence environmental health decisions. This review also underscores current knowledge gaps and identifies future research trends essential for addressing emerging environmental issues associated with CECs. By highlighting areas that require further investigation, this work lays the groundwork for enhanced monitoring and management strategies to mitigate the ecological and health impacts of CECs, thus contributing to more effective environmental governance.

In the context of CECs emanating from biosolids and similarly contaminated matrices like sewage sludge and soil, our review falls within salient research themes about CECs detected in complex media. In addition to the thematic of CECs classification, occurrence, and fate and transport mechanisms covered in this review, additional research thematic areas that have been recently investigated comprise analytical techniques, toxicity, uptake by living organisms, treatment technologies, and risk assessment. This review looked at the occurrence of various classes of CECs in biosolids, sewage sludge, and contaminated soils, providing insight into each class's cumulative occurrence. This work should help provide a framework for data needed to regulate the presence of such CECs for using biosolids for soil enhancement.

# 9.1 CECs' characteristics influencing their fate and transport

Sander et al. (2022) discussed the impact of Henry's Law Volatility Constant  $(H_v)$  on contaminants' environmental fate and migration pathways. The constant plays a critical role in quantitatively influencing the fate and transport of contaminants by determining their distribution between gas and liquid phases within environmental contexts. An elevated  $H_v$  value indicates that the contaminant exhibits an increased rate of evaporation, which is fundamentally significant for predicting the rate at which contaminants can escape water bodies, affecting both water quality and atmospheric pollution levels. This constant is mathematically defined as the ratio of the abundance of the contaminant in the gas phase  $(Q_g)$  to its abundance in the liquid phase  $(Q_l)$  at equilibrium under conditions of infinite dilution:

$$H_{\nu} = \frac{Q_g}{Q_l}$$

Moreover, the constant is a pivotal metric for evaluating the equilibrium distribution of contaminants between air and water, commonly referred to as air-water partitioning. Defined via partial pressure (*p*) of a species in the gas phase relative to its amount fraction (*x*) in the liquid phase under equilibrium conditions at infinite dilution, the Henry's Law volatility constant ( $H_v^{px}$ ) can be expressed as follows:

$$H_{\nu}^{px} = \lim_{x \to 0} \frac{p}{x}$$

The half-life degradation time (DT50) constitutes a significant parameter in elucidating contaminants' environmental destiny and mobility. DT50 represents the time required for the concentration of a specific contaminant to decrease by fifty percent in the environment due to biological and physicochemical degradation





processes. A longer DT50 indicates a persistent contaminant, potentially resulting in extended exposure risks for aquatic ecosystems and human health. In contrast, a shorter DT50 indicates that a contaminant undergoes more rapid degradation, which may mitigate exposure risk. Environmental fate and transport models frequently incorporate DT50 as a key

input parameter to simulate the movement of contaminants through diverse environmental matrices such as soil and water. By integrating DT50 values, researchers can better forecast pollutant concentrations over time in the environment, thereby aiding in assessing their potential impact and residence duration. Furthermore, understanding DT50 can guide remediation



strategies by revealing whether natural degradation processes are adequate or if more proactive treatment measures are warranted (Li et al., 2015).

The organic carbon-water partitioning coefficient (log  $K_{oc}$ ) and the octanol-water partitioning coefficient (log  $K_{ow}$ ) are two additional characteristics that can affect the environmental fate and migratory mechanisms. The log  $K_{oc}$  value aids in forecasting the amount of a pollutant that will remain in the aqueous phase instead of being adsorbed onto soil organic matter. More significant adsorption to soils is correlated with higher log  $K_{oc}$  values. The log  $K_{ow}$  is a crucial metric for evaluating a contaminant's capacity for bioaccumulation. A chemical with a greater log  $K_{ow}$  is more hydrophobic and more likely to partition into organic tissues (USEPA, 1989). The Figure 5 summarizes the fate of CECs during WWT and land application of biosolids and highlights main characteristics that affect their migration in the environment.

### 9.2 Recent advances

Chemometric methodologies have been applied to optimize various parameters within analytical procedures, decipher complex datasets, and interpret results. Prominent examples include the studies conducted by Malvar et al. (2020a), Malvar et al. (2020b), who employed the Box-Behnken experimental design (BBD) to enhance extraction and cleanup techniques. The BBD represents a robust and efficient design that accommodates a quadratic model without necessitating the examination of all potential combinations; it is classified as a form of response surface methodology (RSM) aimed at optimizing processes involving multiple variables. This approach enables the meticulous adjustment of experimental conditions while minimizing the required experiments (Stalikas et al., 2009).

The grouping of contaminants is one of the key strategies proposed by regulatory entities and researchers to control the pollution of CECs (Chirsir et al., 2024; EC, 2006). Recently, advances have expanded beyond BPA to include structural analogs, providing a more holistic understanding of their occurrence in the environment (Huang et al., 2020; Xu et al., 2021a; Xue and Kannan, 2019). Future discussions should also emphasize the importance of permissible thresholds of groups of CECs. For example, bisphenol analogs exhibiting comparable or even greater toxic effects than BPA will compel regulatory bodies to reassess current standards. Specifically, Health Canada's proposed tolerable daily intake (TDI) of 25 µg/kg body weight/day applies exclusively to BPA found in food packaging materials, such as can coatings and lacquers (Health Canada, 2008). Although this proposed TDI is considered conservative, evaluating the risks associated with all bisphenol analogs across all potential exposure pathways is essential. With regards to the permissible concentration of parabens in cosmetics or food additives, the JECFA (Joint FAO/ WHO Expert Committee on Food Additives) instituted an ADI (Acceptable Daily Intake) of 1-10 mg/kg body weight/day applicable to the sum of MeP and EtP (ECCC, 2020). Information on permitted concentrations in the environment and the possible harm they may cause to ecosystems is non-existent, and ADI thresholds do not consider either grouping similar substances or all possible ways of exposure. Thus, there is a need to review permissible intake limits in light of recently acquired information.

During sample preparation, progress was noticed in sample pretreatment procedures, where molecularly imprinted polymers (MIPs) were developed for the sensitive and selective extraction of

BPs from complex matrices. Illustratively, a highly class-selective dummy molecularly imprinted polymer for solid-phase extraction (DMIPSPE) and clean-up enabled to achieve LOQs as low as 0.003 ng/g and 0.0004 ng/g for BPS and BPAF, respectively, during the analysis of BP analogs in wastewater and sewage sludge samples (Sun et al., 2018). As for sample analysis, Matrix-Assisted Laser Desorption/Ionization Mass Spectrometry Imaging (MALDI-MSI) has emerged as an invaluable technique for detecting CECs in complex matrices. Particularly, the MALDI-MSI method facilitated multi-class analysis of 499 organic contaminants within biosolids (Villette et al., 2023). When contrasted with conventional GC-MS and LC-MS techniques, MALDI-MSI offers rapid and minimal sample preparation for analyses and possesses the capacity to identify a diverse range of substances, encompassing CECs. Additionally, Guironnet et al. (2022) implemented a hybrid triple quadrupole-linear ion trap (QqQ-LIT) spectrometer to perform triple-stage mass spectrometry acquisition. Within this

QqQ-LIT framework, Multiple Reaction Monitoring cubed (MS/ MS/MS or MRM<sup>3</sup>) is executed through a series of sequential processes during triple-stage mass spectrometry acquisition. This methodology significantly enhances sensitivity, specificity, and quantification capabilities for complex analyses involving challenging environmental matrices.

### 9.3 Future trends

Antibiotics most frequently identified in sewage sludge or biosolids in this review differ from Canada's five commonly prescribed antibiotic classes. The Canadian Antimicrobial Resistance Surveillance System Report indicates that in 2019, the predominant antibiotic classes utilized in veterinary applications, quantified by weight, were tetracyclines (495,116 kg), macrolides (115,822 kg), penicillins (91,095 kg), sulfonamides (53,226 kg), and lincosamides (46,390 kg) (Public Health Agency of Canada, 2021). On the other hand, the same report lists the defined daily doses (DDD) per 1,000 inhabitants of the most prevalently administered antimicrobials for human use: tetracyclines (1,186.2), extendedspectrum penicillins (1,178.7), macrolides (781.9), penicillin combinations (489.4), first-generation cephalosporins (474.3), and fluoroquinolones (469.3) (Public Health Agency of Canada, 2021). Future research should prioritize the characterization of these antibiotic classes in biosolids and explore the correlation between their usage patterns and prevalence.

This review documents the presence of E2, NRT, NRG, and P in biosolids (Gewurtz et al., 2022). According to Costanian et al. (2018), these hormones are among the most employed in Canada in hormone replacement therapies (HRT). The Canadian HRT products primarily consist of estrogen-only formulations, with estradiol (E2) being a prevalent component (Viglino et al., 2008). These formulations are accessible in multiple forms, including oral tablets, transdermal patches, gels, and vaginal rings. Combined estrogen and progestogen therapies comprise E2 in conjunction with medroxyprogesterone Acetate (MPA), E2 combined with NRT, and E2 paired with NRG (often administered via a patch). Furthermore, formulations utilize combinations of E2 and P that are chemically identical to the endogenous hormones produced by the human body. T therapy is also available to address concerns related to diminished libido or other hormonal imbalances. Future research endeavors should cover this game of hormones and include additional ones such as MPA and others.

Advanced chemometric strategies to process and extract important information from large data sets will receive more attention due to the complexity of modern analytical procedures and instruments and the volume of data generated thereof (Malvar et al., 2020a; 2020b). Multivariate data analysis techniques will be increasingly integrated into chemometric methodologies to allow for the simultaneous exploration of several variables (Shayesteh et al., 2015). Developing chemometric techniques will also prioritize facilitating instantaneous analysis to expedite decision-making. This trend will be particularly significant for environmental monitoring and food safety sectors. Artificial intelligence and machine learning algorithms will play a major role in future data analysis and pattern recognition. Although MALDI-MSI has limitations when quantifying contaminant loads in biosolids, it can be useful for peripheral analysis that could inform traditional LC-MS or GC-MS quantitation assessments (Villette et al., 2023). These methods offer improved quantification, sensitivity, and selectivity for figuring out the quantities of particular substances in complex matrices like biosolids.

Another advancement is the increasing interest in analyzing the degradation byproducts of parent contaminants (Castro et al., 2018; Ma et al., 2018; Malvar et al., 2020b; Riva et al., 2021; Zhu et al., 2019). During the breakdown of contaminants, chemical transformations can lead to the formation of new and unregulated substances. These metabolites may pose equivalent or even higher risks than the parent compounds, as in the case of MeTCS, with a higher bioaccumulative potential than TCS (Coogan et al., 2007), and N-desethylamiodarone more prevalent in sewage sludge than amiodarone (Castro et al., 2018), leading to potentially significant ecological and human health impacts. Researchers have increasingly worked to determine the metabolites and transformation by-products of CECs.

About hormones, it is important to consider the conjugated/ deconjugated process in developing analytical techniques to measure hormones to avoid underestimating their concentrations in the environment (Gewurtz et al., 2022; Naldi et al., 2016). These should have major implications since they underscore the importance of monitoring both conjugated and free forms of hormones in wastewater and soil environments. Future studies on the presence, mobility, and transformation of hormones during WWT and in the environment should consider assessing both conjugated and deconjugated forms of hormones.

While some researchers argue that the persistence of S-DPAs in the environment necessitates further investigations (Lu et al., 2019), others consider that current exposure and treatment technologies sufficiently mitigate risks posed by S-DPAs (ECCC, 2017). Further investigation and observation are necessary to better understand the occurrence and behavior of S-DPAs in the environment. With the recently documented toxicity of 6PPD-Q to coho salmon (Tian et al., 2021; Cao et al., 2022), debates regarding the toxicity levels and acceptable exposure limits to S-PPDs exist, raising questions about the permissible levels of such chemicals in the environment and whether current regulations adequately address these risks. Moreover, the recent determination of the presence of S-DPAs in Arctic seabirds and seals (Lu et al., 2019) highlights the urgency for

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additional research on S-DPAs migration routes and fate in the environment, including the Arctic ecosystem. The persistent of S-DPAs has raised presence concerns regarding bioaccumulation and long-term ecological impacts; therefore, it is essential to better understand how they accumulate in food webs, including Arctic wildlife and Indigenous populations. Public awareness concerning their environmental implications will be indispensable to promote stronger advocacy for sustainable regulatory practices. Regarding S-PPDs, growth in initiatives for tracking their levels in various matrices is expected. More collaborative approaches among environmental scientists and toxicologists to tackle the complexities surrounding S-PPDs and other classes of CECs and their environmental fate will also increase. Klöckner et al. (2021) underscore the need to evaluate the implications of organic contaminants and metal constituents on toxicity and environmental health.

## Author contributions

EH: Conceptualization, Data curation, Formal Analysis, Investigation, Methodology, Validation. Visualization. Writing-original draft, Writing-review editing. and SS: Conceptualization, Funding acquisition, Investigation, Project administration, Resources, Supervision, Validation, Writing-original draft, Writing-review and editing.

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## Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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### Supplementary material

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenvc.2025.1547596/ full#supplementary-material

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