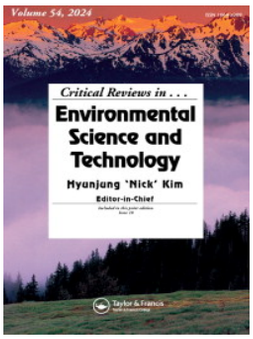

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Triclosan in sludge: Exploring its journey from the sewage treatment plants to land application and potential impacts on the environment

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Triclosan in sludge: Exploring its journey from the sewage treatment plants to land application and potential impacts on the environment

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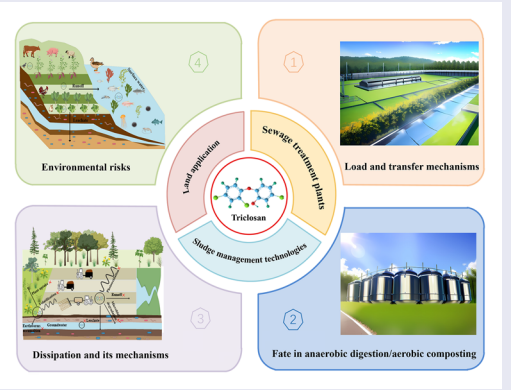
ABSTRACT

Triclosan (TCS) is an anti-microbial widely used in personal care and medical antibacterial products. Despite the widespread occurrence of TCS in municipal sewage sludge, understanding toward the fate of TCS within sewage treatment and environmental risks in the eventual land application is still limited. This review summarizes the TCS loads and transfer mechanisms in the sewage treatment process, sludge management process, land application, and its potential environmental impacts. TCS transfer from sewage to sludge mainly occurs in the primary sedimentation process, representing 2.50 to 4.58 times more compared to the secondary sedimentation process. This transfer is facilitated through adsorption because of the presence of humic acid-like and protein-like substances in sludge. Both anaerobic digestion and aerobic composting contribute to the degradation of TCS with aerobic composting being more effective, exhibiting TCS degradation rates 1.04–2.87 times higher than those observed in anaerobic digestion. After sludge land application, TCS majorly dissipates in the soil through biodegradation by fungi and bacteria, potentially posing environmental risks, such as inhibiting the seedling growth of plant species. Additionally, the degradation of TCS, coupled with the formation and subsequent degradation of MeTCS, is observed, with MeTCS exhibiting a higher half-life and greater toxicity than its parent compound (TCS). Overall, this research offers vital insights to enhance understanding of TCS's migration and degradation processes in sewage treatment and soil. It also provides guidance in environmental protection and sustainable resource management.



KEYWORDS Load and transfer mechanisms; anaerobic digestion; aerobic composting; fate; dissipation; environmental impacts

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1. Introduction

Triclosan (TCS), an endocrine-disrupting chemical, features a halogenated aromatic hydrocarbon composition comprising diphenyl ether, phenol, and polychlorinated biphenyls, endowed with

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antibacterial properties (Ghafouri et al., 2023). TCS has been commonly used in a plethora of routine consumer products, with an average concentration spanning from 0.1 to 0.45% (*w/v*) (Bilal et al., 2020). These products include personal care products and household items (i.e., soaps, toothpaste, shampoo, dish washes) (Weatherly & Gosse, 2017), office and school products (i.e., calculators, adhesives, scissors) (Dhillon et al., 2015), textile (i.e., clothes, bags) (Zhao et al., 2016), and healthcare items (i.e., creams, antiseptics) (Dhillon et al., 2015) for their broad-spectrum antimicrobial properties over a half-century.

The sewage generated from the production, contact, and consumption of products containing TCS is released into domestic sewage systems and downstream sewage treatment plants, which ultimately enters the local environments through effluent discharge or sludge disposal (Dubey et al., 2021; Kulandaivelu et al., 2020; Li, Li, Liu, et al., 2023; Zheng et al., 2020). TCS possesses a high octanol-water partition coefficient (log Kow = 4.76), augmenting its adsorption and precipitation ability in sludge (Blum et al., 2018). Moreover, TCS exhibits greater permeability to lipid membrane, given that most surface waters maintain a pH below 8.1 (pKα of TCS) (Bilal et al., 2020). Consequently, TCS can easily penetrate the aquatic and terrestrial organisms, as well as human body, leading to its accumulation in tissues or organs due to its lipid permeability (Bilal et al., 2020). For example, Baur et al. (2023) reveal that exposure to TCS in concentrations ranging from 0.05 to 0.2% (*w/v*) for 5 d may influence immune regulation by potentially affecting the integrity of the skin barrier and the functioning of human keratinocytes. The swimming behavior of *Pimephales promelas* is markedly impacted by even a short-term exposure of only 1 d to 75 μg/L of TCS (Fritsch et al., 2013). Moreover, after being exposed to 1000 μg/L of TCS for a period of 29 d, *Chlamydomonas reinhardtii* and *Asterococcus superbus* exhibit complete cell collapse (Xin et al., 2019). Therefore, TCS has become a prominent research focus in recent years.

A previous study reports that the concentration of TCS in the influent of sewage treatment plants varies significantly, ranging from 35.6 to 562,000 ng/L (Guo & Iwata, 2017). Numerous review articles have extensively summarized the presence, fate, distribution, removal techniques, and toxicity of TCS in aquatic environments and sewage (Chtourou et al., 2021; Dar et al., 2022; Sun et al., 2023). Specifically, Dar et al. (2022) conclude that the presence of TCS in aquatic environments, coupled with its accumulative properties, lead to genotoxic, neurotoxic, cytotoxic, and hepatotoxic effects on aquatic organisms. Sun et al. (2023) summarize the removal techniques, such as advanced oxidation, biodegradation, and adsorption, under different environmental conditions, emphasizing the potential for enhanced performance through the use of combined techniques. Chtourou et al. (2021) also delve into the removal technologies of TCS from sewage, reaching similar conclusions as Sun et al. (2023).

However, sludge, as a major carrier of TCS in sewage, received limited attention. In previous studies, it is reported that TCS is transferred from sewage to sludge, with approximately 50–64% of TCS accumulating in sludge (Nandikes et al., 2022; Yan et al., 2019), and the predicted accumulated TCS concentration in municipal sludge also exhibits considerable variation, spanning from 13.3 to 30,000 ng/g (Guo & Iwata, 2017). Unfortunately, a thorough summary of TCS loading (mass balance) and transfer mechanisms in sewage treatment is lacking, hampering our comprehension of TCS behavior in sewage sludge plants. Anaerobic digestion and aerobic composting are commonly used techniques to achieve sludge management in sewage treatment plants before final disposal (i.e., land application) (Dubey et al., 2021; Wang, Li, Liu, Zhou, Li, Siddiqui, Lin, Rafe, et al., 2023). However, TCS cannot be completely degraded through these technologies, with anaerobic digestion achieving a degradation rate of 30–46.2% (Narumiya et al., 2013; Wang et al., 2020, 2021; Yang et al., 2017) and aerobic composting resulting in a range of 48.1–86% degradation (Sadef et al., 2014; Yu et al., 2019; Zheng et al., 2020). The degradation extent of TCS during anaerobic digestion and aerobic composting processes has shown significant variation in previous studies (Ying et al., 2007; Yu et al., 2019), creating challenges for a comprehensive understanding of the fate of TCS in different sludge management technologies, and hindering the formulation of corresponding measures to enhance TCS removal. Additionally, a residual amount of TCS will persist in sludge, and subsequently be released into the environment during

biosolids land application. Contradictory conclusions regarding the risks caused by TCS toward environmental or even to human health, have been drawn (Chen et al., 2020; Higgins et al., 2011; Musee, 2018). This inconsistency poses challenges to the development of appropriate strategies to mitigate the associated risks, resulting in knowledge gaps that impede our understanding of TCS behavior in sludge from sewage treatment plants to its subsequent land application.

This review aims to provide a comprehensive analysis of TCS in sewage treatment plants, encompassing its load and transfer mechanisms, degradation during different sludge management technologies (i.e., anaerobic digestion and aerobic composting), fate in agricultural soil following biosolids application, and potential environmental impacts. By examining these aspects, this review offers valuable insights into the behavior of TCS, facilitating informed decision-making to mitigate its environmental impact and contribute to ecosystem safety and environmental well-being.

2. TCS load and transfer mechanisms in sludge during sewage treatment processes

2.1. TCS load in sludge

The sewage treatment plants commonly use the activated sludge method to treat sewage (Hreiz et al., 2015; Liu et al., 2019; Olofsson et al., 2012), where the influent sewage typically passes through primary sedimentation, activated sludge bioreactor, and secondary sedimentation before discharge (Wang et al., 2017) (Fig. 1). In some regions, primary sedimentation tanks are omitted in sewage treatment plants due to specific sewage characteristics, preferred treatment methods, or budgetary and resource limitations, while still ensuring that sewage undergoes proper biological treatment to meet environmental regulatory requirements (Tadsuwan & Babel, 2021; Tchobanoglous et al., 2021; Wang et al., 2019). Additionally, in regions with strict government regulations on sewage discharge, the treatment process commonly includes nitrification and denitrification, essential nitrogen cycling processes in sewage treatment plants (Dai, Gao, Li, Wang, Cui, et al., 2022a; Dai, Gao, Li, Wang, & Duan, 2022). However, the review primarily focuses on sewage treatment plants with the processes of primary sedimentation, activated sludge bioreactors, and secondary sedimentation before discharge, without delving into detailed discussion on TCS load on the nitrification and denitrification processes.

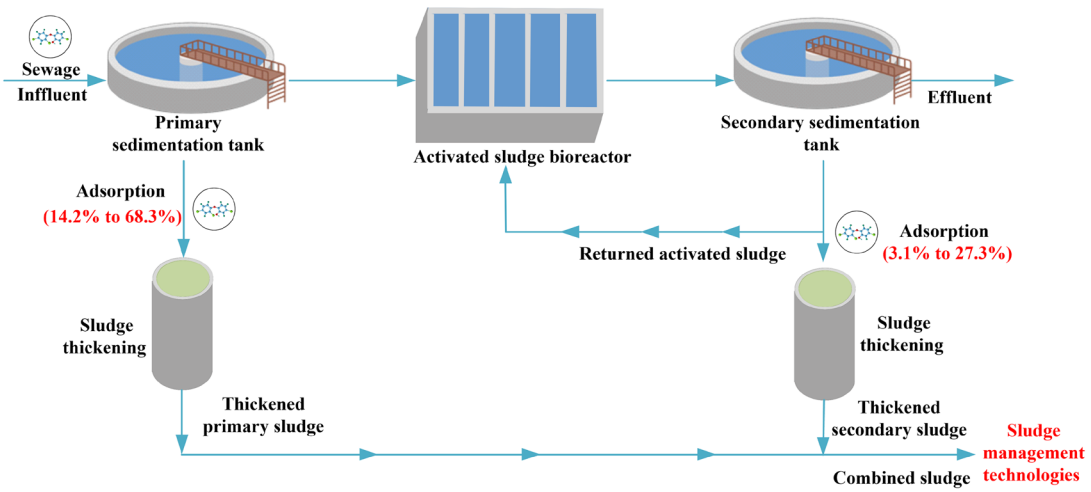

Figure 1. Flow diagram for TCS loading in the solid phase (sludge) of sewage treatment plants. Note: Adsorption represents the main mechanism of TCS transfer from sewage to sludge; (*% to *%) represents the range of TCS in the influent accumulations in the primary and secondary sludge, respectively.

During the sedimentation, part of the TCS is transferred from sewage into sludge, with higher concentrations of TCS observed in primary sludge than in secondary sludge (Lozano et al., 2013; Zheng et al., 2020; Zhou et al., 2020). This difference can be attributed to the greater organic matter content, resulting in increased adsorption surface availability (Elalami et al., 2019; Sakaveli et al., 2021). Typically, between 14.2 and 68.3% of TCS in the influent accumulates in the primary sludge, while 3.1–27.3% is found in the secondary sludge (Lozano et al., 2013, McPhedran et al., 2013; Tohidi & Cai, 2017; Zhang, Wang, et al. 2021). As a result, TCS concentrations detected in the primary sludge are 2.50–4.58 times higher. For instance, the influent of sewage treatment plant, with an average raw sewage flow rate of 1.25 million m^3/d shows that about 68.3% of TCS are present in in the primary sludge, yielding a concentration of 5.74 ± 0.65 kg/d, which is 2.5 times higher than the TCS concentration in the secondary sludge (2.31 ± 0.15 kg/d) (Lozano et al., 2013). Although lower amount (14.2%) of TCS transfer from influent to the primary sludge has been observed in a sewage treatment plant in Hong Kong, TCS concentration in the primary sludge was still 4.58 times higher than that in the secondary sludge (3.1%) (Tohidi & Cai, 2017). The lower transfer rate of TCS can be attributed to rainfall, which elevates the sewage flow rate, consequently reducing the available time for TCS absorption in the sludge. McPhedran et al. (2013) reached similar conclusions, reporting that the accumulation of TCS in primary and secondary sludge is 19 and 3%, respectively. In contrast, a contradictory finding was observed at the Glendale sewage treatment plant in the United States, where the transfer of TCS content to the primary sedimentation was only 7%, notably lower than the 58% removal achieved during secondary sedimentation (McAvoy et al., 2002). This was likely attributed to the high water usage rate reaching 772 L/capital/d (normally 280–537 L/capital/d in other regions) (McAvoy et al., 2002), resulting in a shorter hydraulic retention time in the primary sedimentation tank, consequently limiting TCS adsorption to sludge (Liao et al., 2021; Tohidi & Cai, 2017).

Additionally, the concentration of TCS in primary sludge shows an apparent seasonal variation, with more effective removal during colder temperatures, whereas secondary sludge shows the opposite trend. Tohidi and Cai (2017) find that the reduction of TCS in primary sludge during cold periods (8–16 °C) is 31.17% higher than during warm temperatures (28–36 °C). Similar findings are also observed by Trinh et al. (2016) and Guerra et al. (2019). This is likely due to the higher concentration of TCS in the influent of sewage in cold periods (Awad et al., 2014; Singh & Suthar, 2021; Tohidi & Cai, 2017). On the contrary, TCS concentration in secondary sludge tends to exhibit a lower removal percentage in winter compared to summer, this phenomenon is possibly attributed to reduced microbial activity during the cold periods (Hedgespeth et al., 2012), and a detailed explanation will be provided in Section 2.2.

2.2. The mechanisms of TCS transfer from sewage to sludge

The accumulation of TCS in sludge is majorly through adsorption due to the strong hydrophobic properties of TCS (Fan et al., 2020; Lozano et al., 2013; Tohidi & Cai, 2017). The interaction between sludge and TCS is primarily governed by hydrophobic interaction, hydrogen bond, and electrostatic interaction (Cui et al., 2022; Yan et al., 2019). The organic fractions of sludge commonly consist of 60–80% extracellular polymeric substances (EPS), 15–42% humic substances, 1–6% nucleic acid, and 1–2% uronic acid (Wang, Li, Liu, Zhou, Mou et al., 2023). EPS, macromolecular substances secreted by bacteria, contain majorly proteins and polysaccharides (Chetty et al., 2021; Wang et al., 2014). EPS plays a key role in the adsorption of micropollutants such as TCS to sludge, where the proteins provide the primary adsorption sites (Li et al., 2016; Xiao & Zheng, 2016). A higher ratio of proteins/polysaccharides enhances the availability of adsorption sites and binding ability between EPS and micropollutants (Yan et al., 2021; Zhang et al., 2018). EPS can further be divided into three categories: tightly bound EPS, loosely bound EPS, and soluble EPS (Sheng et al., 2010). Recent research has shown that TCS is primarily adsorbed to tightly bound EPS (50.65%), followed by loosely bound EPS (30.54%) and soluble EPS (18.81%)

(Cui et al., 2022). The TCS content in these three types of EPS is highly associated with their organic substances content (729, 327, and 236 mg/L) and composition (proteins/polysaccharides: 2.79, 2.31, and 2.01) in tightly bound EPS, loosely bound EPS, and soluble EPS, respectively (Cui et al., 2022), with humic acid-like and protein-like matters being identified as crucial players in the interaction between sludge and TCS (Cui et al., 2022; Li, Li, Han, et al., 2023). This finding aligns with previous research that found humic acid can remarkably disrupt the adsorption of TCS in activated carbon due to the higher affinity between TCS and humic acid (Behera et al., 2010).

Additionally, a small portion of TCS is also degraded during the secondary sedimentation. A previous study finds that the total mass of TCS in the influent is comparable to the combined of the primary sludge and primary effluent (Fig. 1), indicating that TCS present in the primary sludge is not eliminated, merely attaches itself to the primary sludge, owing to extremely low presence of microbes (Lozano et al., 2013). However, a significant difference is observed between the total TCS content in the returned activated sludge and primary effluent and the combined TCS content in the secondary effluent and secondary sludge (Fig. 1). This observation, coupled with the negligible impact of TCS reduction owing to photodegradation and volatilization (Tohidi & Cai, 2017), strongly indicates the occurrence of TCS degradation (around 33%) during the activated sludge bioreactor (Lozano et al., 2013). Degradation plays a crucial role in reducing TCS, and variations in reduction pathways can be linked to differences in hydraulic and solid retention times, as well as variations in TCS concentration due to differences in the served population and types of sewage being treated (McPhedran et al., 2013). These aspects need to be further clarified in future studies.

3. The fate of TCS in different sludge stabilization technologies

Sludge stabilization technologies are critical in removing TCS from sludge before its land application, representing the last chance to mitigate or prevent TCS entry into the environment (Abbott & Eskicioglu, 2020a). Anaerobic digestion, aerobic composting, and aerobic digestion are the commonly used sludge stabilization technologies (Martín et al., 2015; Wang, Li, Liu, Zhou, Li, Siddiqui, Lin, Huang, et al., 2023). Anaerobic digestion not only enables energy recovery from sludge but also reduces its volume and enhances its quality compared to raw sludge (Wang, Li, Siddiqui, et al., 2023; Zhang, Li, et al. 2021, Liu et al., 2023). On the other hand, aerobic composting converts organic substances into humic substances and inactivates pathogens present in the sludge, making the treated sludge more suitable for soil improvement and fertilizer production (Balaganesh et al., 2022; Tambone et al., 2010). Due to the higher operating costs and limited usage (Abbott & Eskicioglu, 2020b), aerobic digestion is not discussed in this review.

3.1. The fate of TCS in the anaerobic digestion

3.1.1. Degradation efficiency of TCS in the anaerobic digestion

Conflicting results are reported regarding the removal ratio of TCS in anaerobic digestion (Table S1). Some studies find that the removal ratio of TCS is negligible (non-biodegradable) in anaerobic digestion (Table S1). For example, one study observed that the concentration of TCS does not remarkably decrease at ambient temperature (22 °C) within 70 d anaerobic conditions (Ying et al., 2007). Other studies conducted by Chen et al. (2011), Abbott and Eskicioglu (2020a), and Pycke et al. (2014) also confirm that TCS remains almost unchanged at ambient temperatures ranging from 17 to 25 °C during anaerobic digestion of 25–30 d for combined sludge (primary and secondary sludge) and secondary sludge, respectively. This negligible degradation efficiency in anaerobic digestion conditions might be attributed to the limited microbial activity at ambient temperature, which negatively impacts anaerobic digestion performance and TCS degradation (Liu et al., 2016).

In contrast, Gonzalez-Gil et al. (2016) report a TCS degradation rate of 20.0% in combined sludge (initial concentration of 1.4 mg/kg) under mesophilic anaerobic digestion (37 °C) for a duration of 20 d. Similarly, Samaras et al. (2013) and Tohidi and Cai (2017) also report a comparable degradation rate of TCS (22.0–23.0%) in mesophilic anaerobic digestion (35–36 °C, 17–19 d), with initial TCS concentration of 1.74–3.39 mg/kg. Higher TCS removal rates of 30–46.2% are also found in other studies (Li et al., 2021; Narumiya et al., 2013; Wang et al., 2020, 2021; Yang et al., 2017).

3.1.2. Factors affecting the degradation efficiency of TCS in the anaerobic digestion

Several potential factors can contribute to the variations in TCS degradation efficiency in anaerobic digestion, including the initial concentration of TCS in the sludge, temperature, sludge retention time, etc. (Table S1).

Higher TCS concentration commonly leads to lower TCS degradation rates. For example, in mesophilic anaerobic digestion (35 °C, 20 d sludge retention time), the degradation rate of TCS in combined sludge is 37.3% with an initial TCS concentration of 3.96 ± 1.59 mg/kg (Abbott & Eskicioglu, 2020a), but 46.2% under 2.21 ± 0.55 mg/kg in combined sludge of TCS (Samaras et al., 2014) and 55.0% under 1.97 ± 1.17 mg/kg in the primary sludge (Yang et al., 2016). A significant increase in TCS degradation rate is observed when TCS concentration is reduced to 1.67 mg/kg in the secondary sludge, reaching up to 74.0% under 20 d mesophilic anaerobic digestion conditions (Zhou et al., 2017). This is possible because microbes may have low tolerance to high concentration of TCS, leading to toxicity, reduced microbial activity, and consequently diminishing the overall degradation efficiency of TCS (Wang et al., 2020). However, in some studies, low concentrations of TCS are even close to (or below) the detection limit of TCS in sludge (Ying et al., 2007).

Thermophilic anaerobic digestion (55 °C) is found more efficient than mesophilic anaerobic digestion (35 °C) for TCS removal in sludge. TCS degradation rates vary between 46.0 and 50.0% in mesophilic anaerobic digestion, but 62.0–74.0% in thermophilic anaerobic digestion (Table S1) (Samaras et al., 2014; Zhou et al., 2017). Samaras et al. (2014) report that as increase of the temperature for anaerobic digestion (sludge retention time of 20 d) from 35 to 55 °C increases the TCS degradation rate in combined sludge from 46.2 to 62.0%, with the initial TCS concentration of 2.21 ± 0.55 mg/kg. The findings of Zhou et al. (2017) further support the positive impact of temperature on TCS degradation during anaerobic digestion, where the TCS degradation rate increases from 50.0 to 74.0% when temperature increases from 35 to 55 °C. Also, limited degradation of TCS has been observed at low temperatures (below 25 °C, ambient temperature) (Chen et al., 2011; Pycke et al., 2014; Ying et al., 2007). The heightened microbial activity in the thermophilic environment provides a conducive setting for the efficient degradation of TCS, explaining the observed higher degradation rate in thermophilic anaerobic digestion (55 °C) compared to its mesophilic counterpart (35 °C) (Shin et al., 2019; Wang et al., 2020). This suggests the important role of an optimum temperature in the degradation of TCS during anaerobic digestion.

A longer sludge retention time contributes to TCS removal during anaerobic digestion (i.e., 20–30 d) (Feng et al., 2019; Lee et al., 2011; Ponsá et al., 2008). Under mesophilic anaerobic digestion, increasing sludge retention time from 21 to 30 d and eventually to 65 d leads to an increase in TCS degradation rate (initial TCS concentration of 2.91 ± 0.15 mg/kg dry weight) from 29.8% to 35.7% and 45.0%, respectively (Wang et al., 2021). Yang et al. (2016) report that the degradation rate of TCS in primary sludge, with an initial TCS concentration of 1.97 mg/kg, increases from 46.0 to 58.0% as the sludge retention time increases from 15 to 30 d. Moreover, Zhou et al. (2017) observe an increase in TCS degradation in the anaerobic digestion of secondary sludge (initial TCS concentration of 1.67 mg/kg) from 44.0% to 50.0% and 74.0% as the sludge retention time is extended from 10 d to 15 d and 20 d in mesophilic anaerobic digestion conditions, respectively. Similarly, extending the sludge retention time in thermophilic anaerobic

environments leads to a significant increase in TCS degradation from 52.0 to 72.0%, when the sludge retention time is increased from 7 to 15 d (Zhou et al., 2017). The potential reason can be ascribed to the adaptation of microbial communities to the presence of TCS, further enhancing the efficiency of TCS degradation (Yang et al., 2016).

3.1.3. Impact of sludge pretreatment techniques on the degradation efficiency of TCS in the anaerobic digestion

Various sewage sludge pretreatment techniques have been developed to enhance the energy recovery from anaerobic digestion, including thermal pretreatment, microwave pretreatment, ultrasonic pretreatment, enzymes addition, etc. (Kor-Bicakci et al., 2020; Zhou et al., 2017). The application of sludge pretreatment techniques also shows positive impacts on TCS degradation during anaerobic digestion. For instance, Yang et al. (2017) report that thermal pretreatment (treatment condition of 150 °C and 500 kPa for 30 min) increased TCS degradation rates from 39.0% in the control group (without thermal pretreatment) to 49.0% during anaerobic digestion. Ultrasonic pretreatment (0.05 W/mL energy density for 30 min) enhances TCS removal rate from 50.0% in the control to 73.0% in mesophilic anaerobic digestion of secondary sludge (Zhou et al., 2017). Furthermore, the addition of lysozyme and papain (both at 30 mg/g total solids under the same conditions), key enzymes in promoting sludge anaerobic digestion performance, also increases TCS removal rate from 50.0% in the control to 60.0 and 82.0%, respectively (Zhou et al., 2017). These results indicate that lysozyme and papain positively impacted TCS removal, with papain demonstrating higher efficiency than lysozyme in enhancing TCS removal from the sewage sludge. Additionally, microwave pretreatment (temperature: 160 °C; time: 30 min; frequency: 1200 W) also improves the TCS removal rate from 21.0% in control to 57.6% in thermophilic anaerobic digestion (55 °C, 12 d) of combined sludge (Kor-Bicakci et al., 2020). These observations suggest that the pretreatment of sewage sludge before anaerobic digestion is more efficient than the traditional anaerobic digestion process for degrading TCS.

3.1.4. Degradation pathways of TCS in the anaerobic digestion

TCS degradation in sewage sludge anaerobic digestion possibly involves four pathways: hydroxylation, dechlorination, cleavage of ether bonds (C–O cleavage), and methylation (as illustrated in Fig. 2) (Wang et al., 2020, Wang et al., 2021). Degradation of TCS through the hydroxylation pathway, 5-chloro-2-(2-hydroxy-4-choro phenoxy) phenol is produced (Lee et al., 2011). In the dechlorination pathway, TCS is converted into 2-(2,4-dichloro phenoxy) phenol via reductive dechlorination, with nucleophilic attack on the electron of carbon cations (Chang et al., 2019). The produced 2-(2,4-dichloro phenoxy) phenol is further transformed into 2,4-DCP and 2-(4-dichloro phenoxy) phenol via dechlorination and homolytic reaction of the C-O bond, respectively (Chang et al., 2019). The resulting 2-(4-dichloro phenoxy) phenol can be cleaved into 4-Chlorophenol through ether linkage cleavage. TCS can also be degraded via cleavage of ether bonds (Lam et al., 2019), accompanied by the production of 2,4-DCP and 4-chlorocatechol (Wang et al., 2020). Finally, TCS can be methylated to form methyl triclosan (MeTCS), a typical TCS transformation product frequently observed in sewage sludge and other environmental compartments (Wang et al., 2020). The presence of MeTCS in sewage sludge has raised extensive concerns about its potential environmental impacts. Previous studies have suggested that MeTCS may exhibit similar or even higher toxicity than parent TCS, and it can undergo further transformation and bioaccumulation in organisms (Lozano et al., 2012). Noteworthy, 2,4-DCP is produced in both dechlorination and cleavage of the ether bonds pathway, which may explain why 2,4-DCP is the primary TCS degradation product. This conclusion aligns with previous studies that 2, 4-DCP is an intermediate product of TCS degradation, which can be further degraded to catechol and phenol (Abbott & Eskicioglu, 2020a; Gangadharan Puthiya Veetil et al., 2012; Mulla et al. 2019). Comprehensive explanation of various removal pathways would enhance

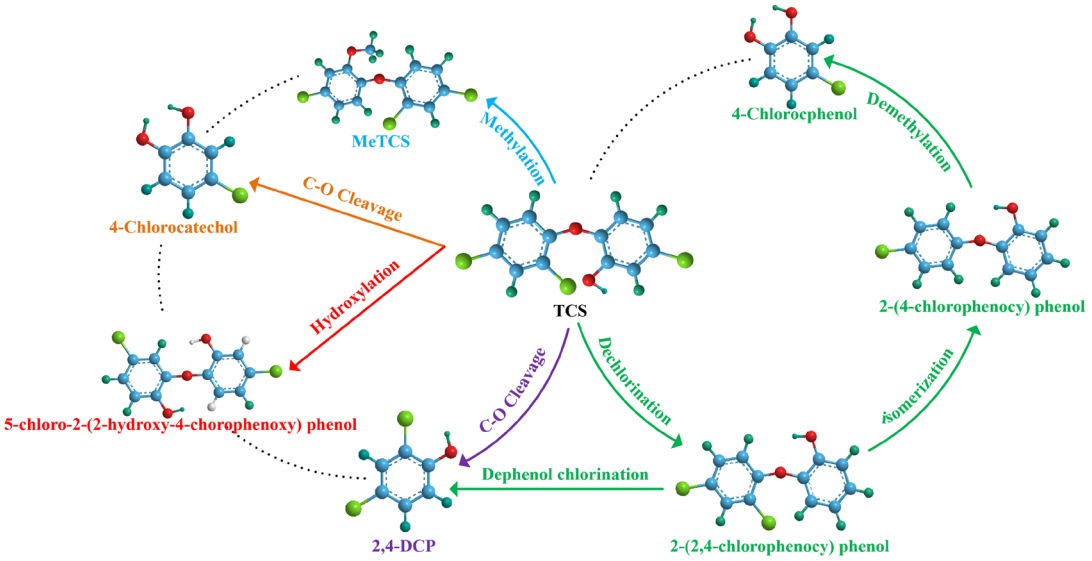

Figure 2. The summarized TCS degradation pathway in sewage sludge anaerobic digestion system based on published articles. The solid arrows represent the direction of substance transformation, with different colors indicating various transformation directions from the parent substance (TCS). Conversely, the dashed lines mean the absence of the specific transformation pathway.

our understanding of the removal mechanisms and routes of TCS during anaerobic digestion processes.

Noteworthy, the microorganisms present within the sewage sludge play a pivotal role in the degradation of TCS during anaerobic digestion, including *Sporosarcina*, *Guggeheimella*, and *Conexibacter* (Wang et al., 2020, 2021). Wang et al. (2020) observe that the relative abundance of *Sporosarcina* is remarkably enhanced due to the elevated TCS exposure concentration (44.7% in the experimental group compared to 0.001% in the control group), indicating that this microbe might be responsible for TCS degradation. This observation aligns with previous research that *Sporosarcina* can degrade some organic contaminants (i.e., TCS) (Li et al., 2018). Moreover, Wang et al. (2021) find that the total abundance of *Guggeheimella* and *Conexibacter* increases along with the increase of TCS concentrations, from 20.1% under 12 ± 2 mg TCS/kg total solids to 29.4% under 322 ± 2 mg TCS/kg total solids, suggesting that these microorganisms might be responsible for TCS degradation in the anaerobic digestion. Understanding the microorganisms responsible for TCS degradation in anaerobic digestion contributes to better managing the removal process of TCS from sludge, thereby reducing its potential environmental impact after digested sludge land application.

3.2. The fate of TCS in the aerobic composting

3.2.1. Degradation efficiency of TCS in aerobic composting

Aerobic composting can also be employed for stabilizing sewage sludge from sewage treatment plants (Karthick et al., 2020; Vasudevan et al., 2021). The aerobic composting of sewage sludge is commonly accompanied by the addition of conditioner or bulking agents, such as sawdust, straw, and returning compost during the composting process, due to the high water content of sewage sludge (70–98%) (Ozaki et al., 2017; Yu et al., 2019). These additives help improve the composting material's structure, porosity, and aeration, facilitating better decomposition and moisture control.

The degradation efficiency of TCS in the aerobic composting process via directly stacked (without the addition of bulking agents) is limited, ranging from 4.1 to 35.0% in previously

reported studies (Ozaki et al., 2017; Yu et al., 2019; Zheng et al., 2020) (Table S2). Zheng et al. (2020) report that the TCS degradation rate during aerobic composting is only 4.1% in 17 d of composting when sewage sludge is directly stacked. Similarly, it is found that the TCS removal rate of sewage sludge is only 17.0% after 16 d of aerobic composting (Yu et al., 2019). The low degradation rate of TCS is primarily attributed to poor aeration (low oxygen concentration), resulting in extremely low microorganism activity in directly stacked sewage sludge (Yu et al., 2019). In other words, the insufficient oxygen supply in aerobic composting is not conducive to TCS degradation (Zheng et al., 2020). Ozaki et al. (2017) observe a TCS removal rate of 35.0% in a 110 d aerobic composting process where sewage sludge is directly stacked. The findings of this study suggest that longer sludge retention time contributes to TCS removal in aerobic composting, which is in line with the conclusion of Armstrong et al. (2018). The addition of conditioner can remarkably enhance TCS degradation efficiency during the aerobic composting process, in contrast to directly stacked (Zheng et al., 2020, Yu et al., 2019). This aspect of the content will be discussed in detail in Section 3.2.2.

3.2.2. Factors affecting the degradation efficiency of TCS in aerobic composting

Several factors influence the degradation efficiency of TCS in aerobic composting, including oxygen content (generated by ventilation) (Yu et al., 2019; Zheng et al., 2020), the ratio of conditioner and sewage sludge, composting temperature, etc. (Table S2).

Increased ventilation during aerobic composting significantly enhances the degradation of TCS in sewage sludge. Yu et al. (2019) observed TCS degradation rates of 54.7, 56.7, and 65.2% for low, medium, and high ventilation treatment, respectively, which are 3.2, 3.3, and 3.8-fold higher compared to the control group (directly stacked; 17.0%). Similarly, Zheng et al. (2020) also note remarkably improved TCS removal rates with increased ventilation during composting, with rates of 48.1, 59.0, and 59.5% for low, medium, and high ventilation treatments, respectively, compared to a mere 4.1% in the directly stacked sewage sludge condition. This phenomenon can be primarily attributed to the increased availability of oxygen within the composting systems, fostering an oxygen-rich environment that supports the growth and activity of microorganisms, including TCS-degrading bacteria (Yu et al., 2019; Zheng et al., 2020).

A suitable ratio of conditioner/bulking agent with sewage sludge positively affects TCS degradation efficiency. A previous study finds that the degradation efficiency of TCS during a 30 d composting period increases from 55.0 to 81.0% when the ratio of sawdust to sewage sludge increases from 1:2 to 1:3 (*v/v*) (Haiba et al., 2017). The potential reason can be ascribed to the following aspects: (1) improved aeration resulting from the increased porosity of the composting mixture (Zhang et al., 2022); (2) balanced carbon-to-nitrogen ratio due to the carbon-rich sawdust (Zhang et al., 2022); (3) enhanced microbial activity as the carbon content from sawdust provides a more abundant energy source for microorganisms, thus promoting their activity (Zhou et al., 2014). Notably, Poulsen and Bester (2010) report a high TCS removal rate of 84.0% during a 24 d composting period when using a composting mixture comprising conditioner materials, such as yard and park waste, straw/house manure, cardboard, and troltex at a ratio of 30:12:7:9, along with the sludge at the ratio of 58:42. The potential reason for this significant removal might be attributed to the sludge being subjected to anaerobic digestion before undergoing aerobic composting (Poulsen & Bester, 2010).

Additionally, selecting the appropriate temperature is crucial to maximize the TCS removal rate. Under varying temperatures of 18, 30, 37, and 70 °C, and employing a blend of 49% conditioner (consisting of 4% straw, 20% yard waste, 10% troltex, and 15% compost residue) mixed with 51% sewage sludge (*w/w*), TCS degradation rates are observed as 58.0, 86.0, 78.0, and 64.0%, respectively (Sadef et al., 2014). Higher temperatures (i.e., 30, 37, and 70 °C) increase TCS degradation rates compared to 18 °C as elevated temperature enhanced microbial activity, boosting TCS degradation. Yet, extremely high temperatures like 70 °C can hinder microbial activity, affecting overall composting. This aligns with the research of Armstrong et al. (2018),

supporting the idea that a suitable temperature is advantageous for TCS degradation. Therefore, to achieve maximum TCS degradation efficiency in sewage sludge, optimizing these factors to obtain optimal results, thereby mitigating its environmental impact during subsequent applications, is necessary.

3.2.3. TCS degradation mechanisms in the aerobic composting

The aerobic composting process of sewage sludge typically consists of three stages: the mesophilic phase (short duration time, i.e., 2 d), followed by the thermophilic phase (longer duration time, i.e., 10 d), and finally, the cooling phase (short duration time, i.e., 3 d) (Yu et al., 2019; Zheng et al., 2020). TCS degradation mainly occurs during the mesophilic phase and early stage of the thermophilic phase, likely due to the higher activity and reproduction of aerobic microorganisms in the mesophilic phase, which results in a remarkable enhancement of the removal rate of TCS (Yu et al., 2019; Zheng et al., 2020). Specifically, *Alkalilimnicola*, *Tissierella*, *Lascolabacillus*, and *Pusillimonas* are known to play a crucial role in TCS degradation, as the degradation rate of TCS has shown a strong link with the relative abundance of these microorganisms (Zheng et al., 2020). Besides, previous studies have reported that *Pseudomonas*, *Sphingomonas*, and *Novosphingobium* in sewage sludge can also degrade TCS (Mulla et al., 2016; Thelusmond et al., 2019). Zheng et al. (2020) find that the relative abundance of *Pseudomonas* in 0th d, 2nd d (mesophilic phase), 7th d (thermophilic phase), and 17th d (cooling phase) is 0.1, 0.5, 2.2, and 0.4% (the ratios in the total microbial community), respectively, in the medium ventilation composting system with 13.9% oxygen content. This correlated with a 58% TCS degradation rate, indicating that aerobic composting process benefits *Pseudomonas* enrichment, and enhanced TCS degradation rate.

4. The fate and potential environmental impacts of TCS in agricultural soil following land application of biosolid

4.1. TCS dissipation in the soil after biosolids land application

Incineration, landfill, and land application are the primary treatment and disposal methods of treated municipal sewage sludge (biosolids), among which land application is more appealing, owing to its potential to serve as a fertilizer or soil amendment (Yakamercan et al., 2021). Approximately 50, 80, 70, 45, and 80% of biosolids in the United States, Australia, United Kingdom, European Union, and Ireland are applied to land, respectively (Chetty et al., 2021; Healy et al., 2017; Liu et al., 2021; McClellan & Halden, 2010; Wiśniowska et al., 2019, Yadav et al., 2019). Unfortunately, even after being treated using different management technologies, such as anaerobic digestion and aerobic composting, a certain amount of TCS may still persist in the biosolids, introducing TCS to the environment (Nandikes et al., 2022; Radhakrishnan et al., 2023; Robinson, 2020).

Biosolids land application typically involves the release of TCS, degradation of TCS, and the formation and degradation of MeTCS from TCS (Lozano et al., 2010, 2012; Thomaidi et al., 2016). Among these processes, the release of TCS marks the first stage of biosolids land application. A previous study observes that the initial concentration of TCS after the application of biosolids (with TCS concentration in biosolids dry weight of 19.1 ± 3.02 mg/kg) on sandy loam land is 9.5 ± 1.5 μg/kg, with a biosolids application rate of 1.08 kg/m^2 and application depth of 10 cm (equal to 2.05 mg/m^2 soil) (Lozano et al., 2012). Interestingly, TCS concentration in the soil gradually increases and reaches its peak value after 2 months or 8 months of application, with no significant difference between two time points (63.7 ± 14.1 vs. 62.8 ± 13.2 μg/kg), suggesting that TCS is mainly incorporated/assimilated into the soil in this period (Lozano et al., 2012). This conclusion aligns with other research findings with the time span of approximately

2 months (Chen et al., 2020), indicating that the initial step, the TCS release, occurs in the first 2 months.

TCS degradation plays a dominant role in the extended application period of biosolids. TCS concentration in the soil decreased from 63.7 ± 14.1 to 48.2 ± 15.3 μg/kg after 1 year of application, 9.9 ± 2.0 μg/kg after 2 years, and 3.7 ± 0.6 μg/kg after 3 years, corresponding to TCS removal rates of 24.3, 84.5, and 94.2%, respectively (Lozano et al., 2012). The background level of TCS (TCS concentration in soil without biosolids application) is achieved after 3 years of application, suggesting a complete degradation of TCS (Lozano et al., 2012). Furthermore, Lozano et al. (2010) find that biosolids application in previously untreated loamy sand soil (sludge application rate of 0.33 kg/m^2 and application depth of 10 cm) results in a TCS degradation rate of 78.2% after 7–9 months (leaving 45.5 ± 6.8 μg/kg of TCS in the soil), and an increased rate of 96.1% after an extended application period of 16–21 months, reducing TCS levels to 4.3 ± 6.8 μg/kg, comparable to background levels (3.3 μg/kg). Other studies also report varying reductions of TCS concentration in the soil (Butler et al., 2012; Cha & Cupples, 2009). For instance, with a biosolids application rate of 0.73 kg/m^2 and a 10–30 cm application depth, TCS degradation reaches 33.9% after 1 year and 72.9% after 2 years (Cha & Cupples, 2009). Moreover, TCS removal rates of 59.0, 72.0, and 74.0% are observed when biosolids are applied in loamy sand, sandy clay loam, and clay soil, respectively, when using biosolids at a rate of 5 kg/m^2 and a depth of 10 cm (Butler et al., 2012).

TCS degradation, along with the formation and subsequent degradation of MeTCS, occurs throughout the biosolids application process. MeTCS (with a concentration in biosolids dry weight of 0.10 ± 0.01 mg/kg), initially present at a concentration of 0.6 ± 0.1 μg/kg in the soil after biosolids application, increases to 20.0 ± 2.4 μg/kg after 8 months application (Lozano et al., 2012). Subsequently, MeTCS concentration reaches its peak value of 34 ± 6.7 μg/kg after 1 year of biosolids application, then it starts to degrade, resulting in concentrations of 22 ± 4.8 μg/kg after 2 years of application and 12 ± 3.1 μg/kg after 3 years of application (Lozano et al., 2012). The MeTCS concentration in soil after 3 years application is higher than the TCS concentration in soil mentioned above (12 ± 3.1 vs. 3.7 ± 0.6 μg/kg), the potential reason can be attributed to MeTCS being more persistent in soil than its parent compound TCS, as indicated by its higher half-life (the time required for the degradation of a compound reaching 50%) compared to TCS (443 vs. 104 d) (Armstrong et al., 2019; Lozano et al., 2012, 2013; Macedo et al., 2017). However, the half-life values of TCS and MeTCS in the soil may vary depending on diverse factors such as soil type, temperature, variable soil microflora, and field management (Chen et al., 2014; Mulla et al., 2019; Wu et al., 2009). For example, Chen et al. (2014) investigate the half-life of TCS in two different field trial sites, and found that the half-life of TCS was 258 and 106 d, respectively, illustrating the impact of field management on TCS degradation. Thence, to obtain precise and trustworthy data about TCS or MeTCS half-life, it is essential to consider these factors' influence on the results. Notably, the detailed calculation of the half-life of TCS is elaborated in Text S1.

4.2. Factors that influence TCS dissipation in soil post land application of biosolids

TCS dissipation in soil is reported to be affected by several factors after biosolids being applied on land, such as temperature, soil types, frequency of biosolids application, etc. (Butler et al., 2012; Lozano et al., 2010). TCS dissipation rate in summer (30 °C) was 2.8, 4.0, and 3.2 times higher than the TCS dissipation rate in winter (8 °C) when biosolids are applied in loamy sand, sandy clay loam, and clay soil, respectively (Butler et al., 2012). The main reason for this phenomenon might be attributed to low temperature limiting the activity of the microbes, resulting in a low degradation rate of TCS in soil (Butler et al., 2012). Besides, the release-adsorption process is also temperature-dependent, further influencing TCS degradation dynamics in the soil system. This research also finds that the TCS degradation rate is 59.0, 72.0, and 74.0% after

4 months of application of biosolids in loamy sand, sandy clay loam, and clay soil, respectively, under the same sludge application rate and depth in the soil (Butler et al., 2012). Clay soil and sandy clay loam possess higher water retention capacity than loamy sand, which may provide a better habitat for microorganisms, including TCS-degrading microbes, thereby facilitating the decomposition and degradation of TCS (Şeker & Manirakiza 2020).

Additionally, in untreated loamy sand soil, biosolids applied at a rate of 0.33 kg/m^2 and a 10 cm depth achieved 96.1% TCS removal over 16-21 months, resulting in a residual TCS concentration of 4.3 ± 6.8 µg/kg akin to the background TCS levels (3.3 µg/kg) in soil without biosolids application previously (Lozano et al., 2010). However, in soil with a history of biosolids application at the same 16-21 months interval, the remaining TCS concentration was 2.2 times higher than the background level, suggesting that repeated biosolids applications may lead to slight TCS accumulation (Lozano et al., 2010). Hence, it is crucial to control the interval of biosolids application, which can reduce the build-up of TCS concentration in soil. More importantly, when there is a sufficiently long interval between the land applications of biosolids (i.e., 3 years after biosolids are applied in land), the TCS concentration in the soil can reach background levels of TCS in the soil (Lozano et al., 2010). This highlights the potential for environmental remediation and the restoration of soil quality.

4.3. Mechanisms of TCS dissipation in soil receiving biosolids through land application

The possible removal pathways of TCS in soil mainly include runoff, leaching, volatilization, photooxidation, biological accumulation in terrestrial organisms (i.e., plant uptake, earthworms), and biological transformation (Biel-Maeso et al., 2019; Canada & Canada, 2012; Chen et al., 2020; Rivier et al., 2019; Sitterson et al., 2018), as illustrated in Fig. 3. Among these pathways, biological transformation is recognized as the primary reason for TCS removal in soils, with MeTCS being formed as the major degradation product. The bacteria involved might be

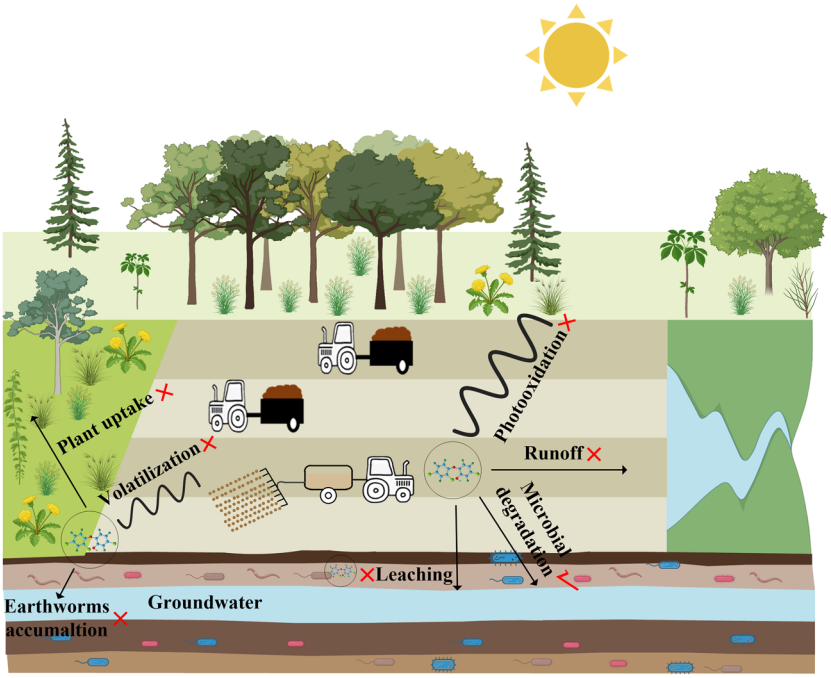

Figure 3. Dissipation pathways of TCS in the soil post biosolids land application. Note: The tick and cross indicate whether the pointed pathways are the primary dissipation pathways of TCS, where the tick represents yes, and the cross represents no.

comparable to those involved in the aerobic composting, as mentioned in Section 3.2.3, various bacteria and fungi are involved in the degradation process, which exhibited good tolerance to the stress caused by TCS, allowing the biodegradation of TCS (Chen et al., 2019). For example, Chen et al. (2020) find that TCS degradation in non-sterile soil (i.e., soil with microorganisms) was 2.16 times higher than in sterile soil during a 56 d incubation period. This observation strongly implies that microorganisms (i.e., bacteria) play a pivotal role in facilitating the reduction of TCS in soil. Previous studies reported that many bacteria can achieve the degradation of TCS, such as *Sphingomonas* sp., *Pseudomonas putida*, *Alcaligenes xylosoxidans, Pycnoporous cinnabarinus* and *Trametes versicolor*, etc. Specifically, Kim et al. (2011) reveal that *Sphingomonas* sp. PH-07 could catabolize TCS into intermediates, including the ether bond cleavage products (i.e., 2,4-dichlorophenol and 4-chlorophenol) and hydroxylated compounds (i.e., dihydroxy-TCS and monohydroxy-TCS), suggesting that the aromatic rings of TCS are the initial sites for dihydroxylation reactions. The findings are consistent with a previous study that reported *Sphingomonas*-like organisms can transform TCS to CO_2 (Hay et al., 2001). Dai et al. (2020) mentioned that *Pseudomonas putida* and *Alcaligenes xylosoxidans* can utilize TCS as a carbon source. Additionally, Hundt et al. (2000) also find that *Pycnoporous cinnabarinus* and *Trametes versicolor* possess the ability to metabolize TCS. *Pycnoporous cinnabarinus* can transform TCS to glucoside conjugate and 2,4,4′-trichloro-2′-methoxydiphenyl ether, while *rametes versicolor* converts TCS into 2-O-(2,4,4′-trichlorodiphenyl ether)-β-D-methoxydiphenyl ether, 2,4-dichlorophenol, and 2-O-(2,4,4′-trichlorodiphenyl ether)-β-D-xylopyranoside (Hundt et al., 2000). By understanding the degradation mechanisms and the involvement of microorganisms of TCS in soil, more effective approaches to manage and reduce the presence of TCS in the environment, thus promoting environmental protection and sustainable development.

Runoff refers to the flow of water that travels over the land surface, typically occurring when water cannot be absorbed by the soil (Sitterson et al., 2018). However, runoff was not considered a significant pathway for TCS dissipation from the soil after biosolids application, removing less than 0.5% of the TCS (Healy et al., 2017) or even less (0.04%) (Sabourin et al., 2009). Leaching is the process of TCS moving from the soil surface into deeper layers as water percolates through the soil (Biel-Maeso et al., 2019). A previous study observed that one year after biosolids application, the concentrations of TCS and MeTCS at a depth of 25 cm were 2.8-fold and 2.7-fold lower, respectively, when compared to their corresponding concentrations at a depth of 10 cm (Lozano et al., 2012). These results suggest that TCS removal through leaching appears insignificant, as the highest TCS concentration remains in the surface soil. Additionally, the removal of TCS in the soil through volatilization and photooxidation is also limited due to the low vapor pressure of TCS in soil (4.65×10^{-6} mm Hg at 25 °C) (Chen et al., 2020) and only a thin layer (several millimeters) located on the surface of soil is affected by sunlight (Canada & Canada, 2012). It is worth noting that although the loss of TCS in soil due to plant uptake was negligible (Hu et al., 2021), a significant amount of TCS is accumulated in the root and leaf of soybeans after applying biosolids to the land (Wu et al., 2010). This may pose a severe threat to human health and the ecosystem.

4.4. Potential environmental impacts of TCS through biosolids land application

Controversial conclusions have been found about the ecological risks posed by the entry of TCS into the environment. Specifically, a preliminary assessment conducted on six terrestrial plant species (e.g., wheat, corn, soybean, ryegrass, tomato, and cucumber) revealed that the calculated risk quotient for these species reached as high as 1360, indicating that these plant species are at significant risk of potential adverse effects for their seedling growth (Ying & Kookana, 2007). Similarly, Chen et al. (2014), Chen et al. (2020), and Abril et al. (2020) have all reported the adverse effects of exposure on terrestrial systems. However, other studies, such as Musee (2018) and Chen et al. (2014), reveal that TCS has no adverse effect on terrestrial organisms.

The potential reasons for these conflicting findings might be the differences of TCS concentration in biosolids in different WWTPs of different countries, the biosolids application rate and application depth, the interval of biosolids application in the same place, etc. Therefore, further research efforts are imperative to conduct a comprehensive global assessment of the potential environmental impacts of TCS following the application of biosolids to land.

5. Future research perspectives

Numerous studies have shown that both primary and secondary sedimentation tanks in sewage treatment plants effectively transfer TCS from sewage to sludge, primarily through adsorption (Tohidi & Cai, 2016). Remarkably, the primary sedimentation tank exhibits superior efficiency compared to the secondary sedimentation tank (Tohidi & Cai, 2016). However, limited research has been focused on monitoring and assessing the effects of MeTCS at various stages of sewage treatment processes. Extensive experimental evidence suggests that MeTCS is more persistent and has higher bioaccumulation potential than its parent TCS, with a log Kow of 5.0 (Lozano et al., 2012; Macedo et al., 2017). Therefore, it is recommended that more targeted monitoring studies be conducted at sewage treatment plants to evaluate the presence and impact of MeTCS at each process of the sewage treatment plants. Furthermore, gaining a comprehensive understanding of the microbial communities within the treatment process that are responsible for TCS degradation is imperative. This knowledge can help identify the specific microorganisms responsible for TCS degradation, particularly in the activated sludge bioreactor (Fig. 1). Such insights can potentially enhance the overall removal efficiency of TCS within sewage treatment plants.

Previous studies have reported that anaerobic digestion can contribute to the degradation of TCS to some extent (Gonzalez-Gil et al., 2016; Narumiya et al., 2013; Samaras et al., 2013, 2014; Wang et al., 2020; Yang et al., 2017). Meanwhile, this technology exhibits longer treatment times, up to 30 d, and is sensitive to pH and temperature fluctuations, resulting in the instability of anaerobic digestion performance (Wang, Li, Liu, Zhou, Qin, et al., 2023). Notably, this process may occur through four pathways, including hydroxylation, dichlorination, cleavage of ether bonds, and methylation) (Wang et al., 2020, 2021). However, the corresponding contribution of each possible pathway for the degradation of TCS remains unknown. Future studies are encouraged to: (1) investigate and quantify the relative contributions of each degradation pathway during anaerobic digestion; (2) reveal the microbial communities and their corresponding metabolic pathways responsible for these degradation processes; (3) develop kinetic models to predict the rates and extents of TCS degradation through individual pathways, helping to elucidate the underlying mechanisms and potential bottlenecks in the process; (4) assess the environmental fate of the byproducts generated from each degradation pathway to determine whether they exert potential risks to ecosystems or human health. Additionally, previous studies found that applying pretreatment techniques can enhance sludge hydrolysis, expedite methane production, reduce digestion time, and positively impacted TCS degradation during anaerobic digestion (Liu et al., 2023; Kor-Bicakci et al., 2020; Zhou et al., 2017). Unfortunately, these technologies often come with the burdens of high energy consumption, elevated treatment costs, and limitation practices, particularly at the laboratory-scale. Consequently, future research could prioritize the exploration of cost-effective and environmentally friendly pretreatment methods to enhance TCS degradation in industrial-scale anaerobic digestion applications in sewage treatment plants.

Studies have indicated that aerobic composting is also conducive to TCS degradation and is more effective than anaerobic digestion in promoting TCS degradation (Yu et al., 2019; Zheng et al., 2020). Notably, aerobic composting also has its limitations, including high energy inputs for aeration, leading to increased operational costs, and the generation of a larger volume of residues compared to anaerobic digestion, requiring more space for disposal (Zhang et al., 2023). Additionally, achieving complete degradation of TCS within sludge during the composting process remains a significant challenge due to its recalcitrant nature. Consequently, more efforts are

required to reduce TCS concentration in sludge before its eventually released into the soil. The mesophilic period during composting is shorter (Yu et al., 2019; Zheng et al., 2020), indicating that enhancing microbial activity during this period seems plausible for enhancing TCS degradation in the composting process. Additionally, few studies have focused on identifying the functional microbes involved at different stages in aerobic composting for a thorough understanding of TCS degradation. Therefore, more research is highly recommended to understand the correlation between specific microbial populations' abundance at different stages and the efficiency of TCS removal during aerobic composting processes. Such efforts are beneficial to a well-designed and sustainable removal of TCS before land application of biosolids. Moreover, some studies reported that TCS removal efficiency could be further enhanced (up to 84.0%) when the sludge is first subjected to anaerobic digestion and aerobic composting (Poulsen & Bester, 2010). However, the relevant studies are extremely limited. Thus, further research to confirm and explore the feasibility of implementing this approach in sewage treatment plants are necessary. More importantly, the degradation of TCS may lead to the production of more persistent byproducts (i.e., MeTCS) that have more severe health impacts (Munaretto et al., 2016). Therefore, future studies are required to identify and characterize TCS's transformation products and determine their potential risk to the environment and even to human health.

Previous studies have shown that the main reason for TCS degradation in agricultural soil following land application of biosolids can be attributed to microbial degradation rather than photooxidation, runoff, or leachate (Chen et al., 2019) (as illustrated in Fig. 3). Unfortunately, most of the current research only performed short-period monitoring (e.g., 1–3 years) (Butler et al., 2012; Lozano et al., 2012) in comparison to the continuous land applications. Thus, long-term monitoring studies are recommended to assess the persistence and accumulation of TCS in agricultural soil, as well as the impact of TCS on soil microbial activity and the potential for TCS to disrupt important soil processes, such as nutrient cycling and decomposition (Lenz et al., 2017). Additionally, controversial observations have been reported regarding the environmental impacts of TCS in following land applications (Musee, 2018; Ying & Kookana, 2007). These discrepancies can be attributed to the regional differences in soil conditions, biosolids

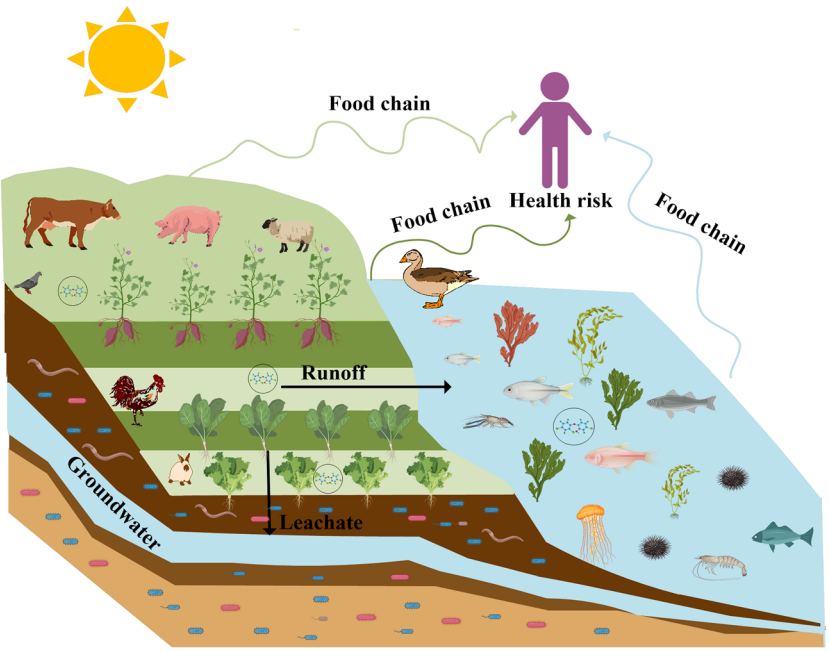

Figure 4. Pathway diagram of TCS entering the human body through the food chain.

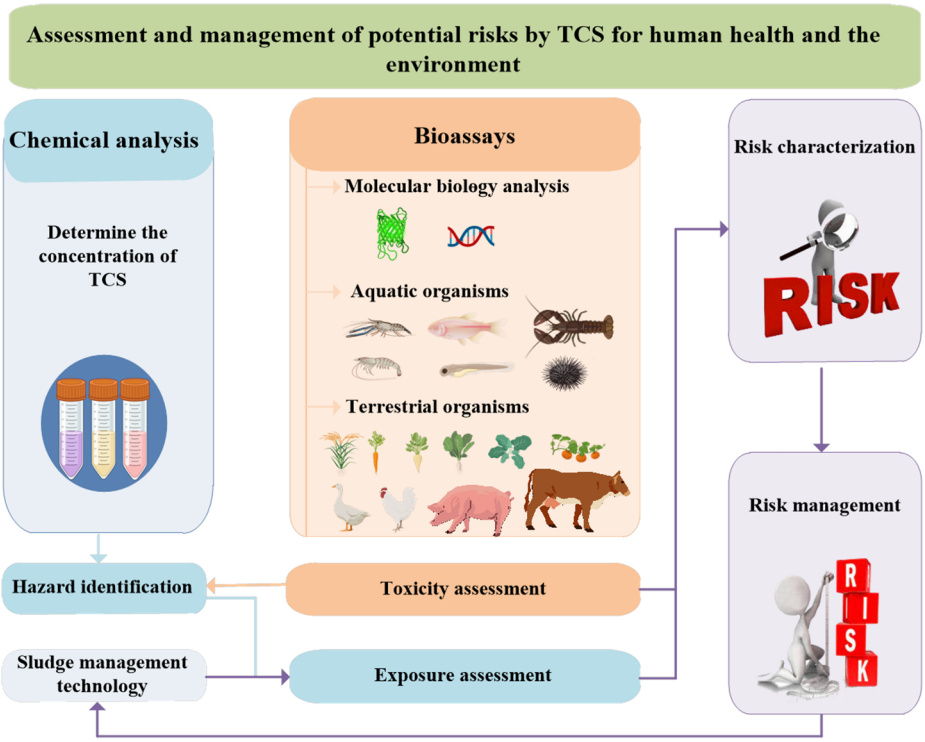

Figure 5. Assessment and management of potential risks posed by TCS to the environment and human health.

application rate, application depth, interval of biosolids application, etc. (Verlicchi & Zambello, 2015). The detailed impact of these factors on TCS degradation is largely unknown, requiring future research. This will help provide a more comprehensive understanding of the overall effects of TCS on the environment (including aquatic and terrestrial compartments). Additionally, the available literature offers limited information on TCS infiltration rate, groundwater contamination potential, adsorption capacity, etc. These data can provide insights into the behavior, transport mechanism, and potential effects of TCS on groundwater quality in soil or water. Therefore, further research on these aspects is highly recommended in future studies.

More importantly, TCS may ultimately accumulate in the human body through food chains (e.g., plants, animals, aquatic, drinking water), posing risks to human health (as shown in Fig. 4). However, limited studies focused on the toxicity of TCS in various environmental compartments and its potential impacts on the ecosystem, even to human health. Thus, for future studies, accurate assessment and management of potential risks caused by TCS for the environment and human health are needed, as illustrated in Fig. 5. Additionally, there is a need to understand the toxicological effects of TCS on wildlife species, such as mammals, birds, etc. through the application of multiple omics techniques (e.g., proteome and transcriptome). This can help identify potential biomarkers and clarify toxic mechanisms. In this way, a more precise characterization of the risks posed by TCS can be achieved, enabling the development of corresponding strategies for effective management.

6. Conclusions

This study provides a critical review of TCS in sludge: exploring its journey from the sewage treatment plants and potential environmental impacts. The essential conclusions of this review are concluded as follows:

1. The transfer of TCS from sewage to sludge primarily takes place in the primary sedimentation tank of sewage treatment plants, showing 2.50–4.58 times higher than the secondary sedimentation tank. The transfer is primarily attributed to the adsorption of TCS onto sludge particles with the process predominantly influenced by the presence of humic acid-like and protein-like substances in the sludge.
2. Anaerobic digestion and aerobic composting all contribute to the degradation of TCS present in sludge, with distinct groups of microbes in charge of the degradation process. However, aerobic composting has demonstrated a higher effectiveness in TCS degradation than anaerobic digestion, with enhancements ranging from 1.04-fold to 2.87-fold.
3. Microbial degradation is the primary factor driving the dissipation of TCS in soils, with MeTCS being formed as the major degradation product-a more persistent byproduct than parent TCS. Moreover, the land application of biosolids could potentially present significant risks to terrestrial systems or even to human health via food chains.

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