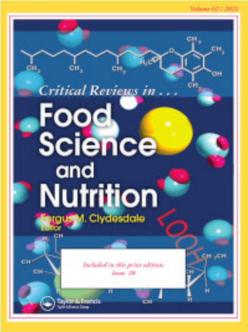

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REVIEW

Chemical food safety hazards in circular food systems: a review

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ABSTRACT

Food production has increasingly become effective but not necessarily sustainable. Transitioning toward circular production systems aiming to minimize waste and reuse materials is one of the means to obtain a more sustainable food production system. However, such a circular food production system can also lead to the accumulation and recirculation of chemical hazards. A literature review was performed to identify potential chemical hazards related to the use of edible and non-edible resources in agriculture and horticulture, and edible plant and animal by-products in feed production. The review revealed that limited information was available on the chemical hazards that could occur when reusing crop residues in circular agriculture. Frequently mentioned hazards present in edible and non-edible resources are heavy metals, process and environmental contaminants, pesticides and pharmaceuticals. For feed, natural toxins and pharmaceutical residues are of potential concern. Studies, furthermore, indicated that plants are capable of taking up chemical hazards when grown on contaminated soil. The presence of chemical hazards in manure, sewage sludge, crop residues, and animal by-products may lead to accumulation in a circular food production system. Therefore, it is relevant to identify these hazards prior to application in food production and, if needed, take precautionary measures to prevent food safety risks.

KEYWORDS

Chemical hazard; circular; agriculture; horticulture; feed

Introduction

After World War II, governments focused on maximizing yields of food production, which led to the development of new technologies, mechanization and increased use of chemicals. Since then, plant and animal production systems in Europe have become highly efficient providing safe and high-quality food for everyone. However, this has resulted in some negative side effects: our current feed and food production systems have impacted our planet by, for example, soil deterioration and the emission of greenhouse gasses. These side effects together with the growing world population and the limited availability of resources have led to an increased need for sustainability. One way to obtain a more sustainable production system is to make it more circular. Within circular agriculture, waste is minimized and resources are reused as much as possible (Hamam et al. 2021). Closing loops to make our food production system more circular will lead to less food waste thereby contributing to more sustainable use of natural resources (Diacono et al. 2019).

With the establishment of the Green Deal and Farm to Fork policy, the European Commission is aiming for a green transition toward 2030 (European Commission 2021). An important part of the proposed way forward is to reuse the by-products from food production, processing and consumption into the food production system. By-products are secondary products obtained during the manufacturing of other products. In order to avoid waste, these by-products should be re-introduced into the food supply chain at the highest level possible (human food – animal feed – soil fertilization). By-products are frequently used as animal feed. These include plant-based products, or crop residues, like skins, peels, seeds, or sugar beet pulp and animal-based products from the dairy, meat and fish processing industry such as whey and milk powder, beef, pork and poultry fat, fish meal, and fish oil (Ominski et al. 2021; van der Fels-Klerx et al. 2019). In addition to reusing by-products, the reuse of waste streams also contributes to the circularity in agriculture. Examples are the use of treated waste water and application of biosolids from municipal waste in agriculture, which is increasing due to their nutrient content, their considerable economic benefits and contributions to waste minimization (Hamam et al. 2021). Replacing artificial fertilizers by the application of animal manure is another example of practices to increase circularity. However, such circular food production systems do not only have advantages, but also entail disadvantages and tradeoffs. When by-products are reused repeatedly over a long period of time or at increased frequency, food safety hazards could accumulate or recirculate in the food chain (Lopes et al. 2011). For example, compounds of emerging concern (CECs), such as pharmaceuticals and personal care products, end up in small amounts in the environment, but could accumulate when taken up by plants. These plants are subsequently reused as

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organic amendments for crop production (Goss, Tubeileh, and Goorahoo 2013; Mansilla et al. 2021). Composting or anaerobic digestion can be applied to reuse food waste in circular food systems. However, such procedures not necessarily reduce food safety hazards. A recent review showed that organic compounds such as polychlorinated biphenyls (PCBs) and perfluorinated compounds are persistent and are not affected by composting or anaerobic digestion (Thakali and MacRae 2021). This may result in accumulation of these compounds in the environment and subsequent contamination of final food products resulting in human health risks (Goss, Tubeileh, and Goorahoo 2013).

Although the EC Green Deal and Farm to Fork policy advocate the closing of loops in our food production systems, they do not provide policies to monitor and ensure the safety of by-products. Nevertheless, it is the responsibility of the food business operator to put safe products on the market (EU Regulation (EC) 178/2002). Therefore, when using byproducts within a food supply chain, potential food safety hazards should be taken into account. So far, however, little is known on food safety hazards when closing the loops. Available data on possible food safety hazards and their fate in circular agriculture are limited and scattered. This review aims to provide an overview of food safety hazards that may occur when closing loops in the food production system. The focus of the study was on chemical hazards. Figure 1 shows that multiple routes are possible when closing the loops in agriculture. Since not all routes could be evaluated, the study focused on a selection of these routes: the application of by-products and waste streams as fertilizers for agriculture and horticulture and as ingredients for feed production. This includes non-edible streams from wastewater treatment plants (e.g. sewage sludge and other sludge types) and animal manure, and edible resources from crop residues and animal by-products (indicated in the colored blocks in Figure 1).

Methods

A systematic literature review was performed to identify the possible presence of chemical hazards related to closing the loop in agriculture and horticulture and in feed production. Three separate searches were performed for which specific search terms were established as indicated in the Annex, being: (1) Chemical hazards related to non-edible sources in agriculture and horticulture; (2) Chemical hazards related to the use of crop residues in agriculture and horticulture or feed production; (3) Chemical hazards related to the use of animal by-products in feed production. Scopus and CAB were used to search for relevant papers published in the English language in the period after 2010. The obtained references were downloaded in three separate Endnote files (one each for the separate searches) and duplicates were removed. A two-tier approach was used for evaluation of the relevance of the retrieved papers. First, the references were screened on title, keywords, and abstract. A second expert reviewed 10% of the papers to check for consistency. Inconsistencies were discussed and the evaluation was aligned. References that were evaluated as relevant or possibly relevant in the first tier were read in full in the second tier to arrive at a final selection of relevant papers. This selected set of relevant papers was used to draft this review. For all searches, snowballing was applied while reading the full texts of the relevant articles. Furthermore, information from previous research on chemical hazards in various food and feed supply chains (Banach et al. 2019; Hobe et al. 2020; Hoffmans, Hoek-van den Hil, and van Asselt 2020; Klüche, Hoek-van den Hil, and van Asselt 2020; Nijkamp

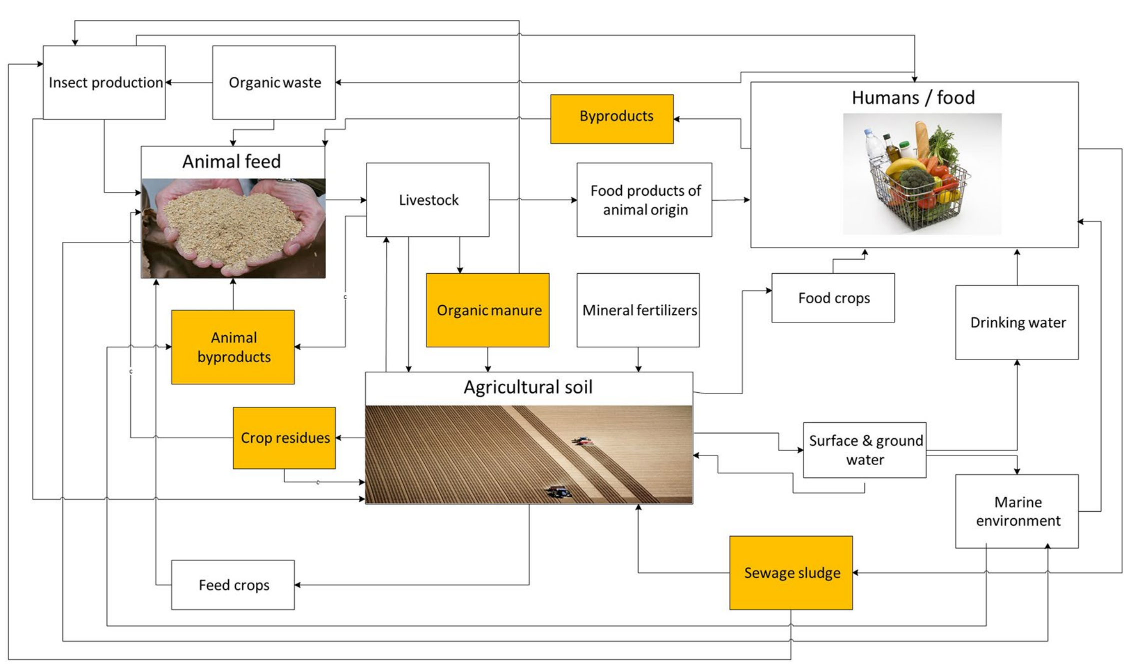

Figure 1. Illustration of the use of side streams and by-products thereof in circular food production systems.

et al. 2017; van der Fels-Klerx et al. 2019) was used to supplement the information obtained.

Results

The literature search on non-edible sources in agriculture and horticulture resulted in 184 hits of which 143 references were considered relevant or maybe relevant based on title, keywords and/or abstract (tier 1). After reading the full texts (tier 2), 25 hits were judged relevant. The search terms on edible resources, that is, crop residues and food by-products, in agriculture and horticulture and feed production resulted in 249 hits. Around 50 of these were on biochar applications. Information on biochar in this paper was added from obtained review papers instead of adding the individual papers on biochar. In the first tier, 40 of the papers not describing biochar were seen as relevant. The second tier resulted in a final section of 15 relevant hits. The literature research on the use of animal by-products in feed resulted in 21 papers of which 2 were relevant to include. Apparently, in recent years not much research on the presence of chemical hazards in animal by-products to be used in feed has been performed. However, animal by-products have been used in feed for many years and their associated chemical hazards are known. Therefore, for this study, a recent overview of the feed supply chain was used to describe the relevant chemical hazards related to animal by-products (van der Fels-Klerx et al. 2019). All relevant information was gathered and summarized in the sections below in which the results were restructured to the use of non-edible resources in agriculture and horticulture (section "Hazards related to the use of non-edible resources in agriculture and horticulture"), edible resources in agriculture and horticulture (section "Hazards related to the use of edible resources in agriculture and horticulture"), and edible resources in feed (section "Hazards related to the use of edible resources in feed").

Hazards related to the use of non-edible resources in agriculture and horticulture

Interest in using both animal and urban waste as soil amendments in agriculture and horticulture is growing nowadays since it enables enhancing the soil quality and, simultaneously, contributes to a sustainable way of waste management. Organic material application such as sewage sludge and animal manure either raw or as compost are well-known practices around the world since their use as fertilizer provides nutrients (e.g., nitrogen, phosphorus and sulfur) to the soil and increases soil organic matter (SOM) (Celik et al. 2010; Tejada et al. 2008). According to the European Commission (2006b), classification of organic waste and compost is based on the origin of its raw materials: agricultural and forest residues, urban residues, wastewater treatment sludge, residues from industrial processes, and their mixtures. The use of the aforementioned animal and urban waste reduces the application of artificial fertilizers. Concerns exist about the potential migration to the food chain of inorganic (heavy metals) and organic (e.g. pesticides, pharmaceuticals and personal care products) pollutants that are present in animal and urban waste. Thus, food safety hazard characterization of these waste types is needed to reduce any possible undesired effects and to regulate and encourage their use in sustainable agriculture. The following sections describe the possible chemical hazards related to both animal and urban waste used in agriculture and horticulture.

Heavy metals and other elements

Introduction of heavy metals through the use of animal and urban waste (e.g. wastewater sludge, manure) as fertilizer in agriculture has become a potential threat to soil ecosystems and to food safety (Hamuda 2013; Manea, Manea, and Robescu 2013). The EU Fertilizing Products Regulation (EU) 2019/1009 that comes into force on July 16th 2022 states that use of fertilizing products, such as manure, should not lead to exceedances of legal limits in the final feed or food product. EU Directive 86/278/EEC sets rules on the safe use of sewage sludge in agriculture and provides maximum limits for seven heavy metals (cadmium, copper, nickel, lead, zinc, mercury, and chromium) for both the soil and the sewage sludge. However, repeated application of waste as fertilizer in agriculture and horticulture over extended periods of time and at increased frequency favor metal accumulation in the soil and transfer toward the food crops (Lopes et al. 2011). Data related to quantitative determination of heavy metal pollution in agriculture and horticulture due to the use of wastewater sludge and manure are limited in the available literature. Zhang et al. (2011) analyzed 224 animal manures (pig, chicken, and cattle) and feeds to determine copper (Cu) concentration. Pig manures showed clearly higher mean Cu concentration (642.1 mg kg^{-1}) than chicken (65.6 mg kg^{-1}) and cattle (31.1 mg kg^{-1}) manures. Results demonstrated that the Cu concentration in manure was correlated with the Cu concentrations in the animal feed which were 179.8, 20.8, and 16.6 mg kg^{-1}, respectively. Studies on the heavy metal pollution during the application of sewage sludge, mixed municipal solid waste compost and compost produced from agricultural wastes (Alvarenga et al. 2017) showed that the use of sewage sludge caused an increase in the concentration of Nickel (Ni), Zinc (Zn), and Cu in the crops grown on the amended soil, whereas this was not noticeable with compost applications. Besides, Fekri and Kaveh (2013) studied the presence of heavy metals in soil after amendment with solid waste compost and cattle manure and found that the presence of Cd, Pb, Zn, and Cu increased. Results showed that the soil amended with solid waste compost present higher metal content than cattle manure although they highlighted that in both cases the metal content was below the maximum permitted level by the United States Environmental Protection Agency (US Environmental Protection Agency 1993). Paradelo, Villada, and Barral (2020) evaluated the relocation of heavy metals from five metal-rich urban waste composts to crops such as lettuce and ryegrass. Authors found that Zn and Cu were the two heavy metals with the highest concentration in both crops. López-Rayo et al. (2016) and (Martí et al. 2016) determined concentrations of heavy metals in soil and vegetable crops were after long term application of composted sewage sludge (11 years) or manure (13 years) as fertilizer. Results showed that concentrations of heavy metals or elements could increase in pea plants and oat grains (López-Rayo et al. 2016) but estimated hazard quotients (HQs, defined as ingested dose divided by the Reference Dose) showed no human health risks were established (Martí et al. 2016). Lopes et al. (2011) reviewed the heavy metal content of different types of organic waste based on a metareview to estimate possible human health risks derived from the reuse of organic waste and their applications. The authors indicated that highest HQs were obtained for Zn, as compared to cadmium (Cd), Cu, Ni, and lead (Pb) due to the high Zn biotransfer potential. Studies of metal concentration in sludge from several industries show that metals such as silver (Ag), Cu, Pb, and Zn can be present in high concentrations (Bai et al. 2012). Authors demonstrated that the majority of industrial sludge showed high heavy metal concentrations and, consequently, they should not be used as soil fertilizer.

Organic contaminants

A wide range of organic contaminants are found in sewage sludge and waste water, related to consumer and industrial use and practices. These contaminants can subsequently be taken up by crops grown on soil treated with this sludge and waste water. One of these contaminants is the group of per- and polyfluoroalkyl substances (PFAS, a.k.a. "forever chemicals"), which is a chemical class currently under high attention of policymakers and risk assessment agencies. PFAS were found in crops (lettuce and carrot) after the application of compost amended soils. The presence of perfluorooctanoic acid (PFOA) (10–55 ng g^{-1}), 8:2 perfluoroalkyl phosphate diester (8–249 ng g^{-1}) and its degradation products demonstrate that uptake from the environment is possible (Bizkarguenaga et al. 2016). Phenolic compounds are well known emerging hazards that can be present in wastewaters from several industrial, pharmaceuticals and petroleum refineries. Studies on removal of phenol and 2,4-dinitrophenol from industrial wastewater indicated that the use of chicken manure biochar is a promising method to efficiently remove phenolic compounds prior to the application of urban wastewater to the agricultural field (Thang et al. 2019). The accumulation of polybrominated diphenyl ethers (PBDEs) in the soil has been evaluated by Gaylor et al. (2014). The authors investigated soil ecosystems with long-term sludge and manure amendments. They reported PBDE levels from 150 to 6700 µg g^{-1} for $_{3\text{-}7}$PBDEs and 7500 µg g^{-1} for BDE-209 in soil from the sludge amended site. Apart from accumulating PBDEs, sewage sludge was also found to accumulate microplastics (Jiang et al. 2020). When applied to the soil, these microplastics can be taken up by the crops (see section "Plastics and nanomaterials").

Pesticides

Pesticide residues are often found in the soil either due to the direct application of pesticides on land for crop protection or due to their presence in organic waste that is later used as fertilizers. Muola et al. (2021) studied the effect of glyphosate-based herbicide contaminated poultry feed on crop production via poultry manure. They found a decrease in plant growth and vegetative reproduction. Depending on the biodegradability and properties of the pesticides present in organic waste, the biological composting process might either concentrate or reduce pesticide residues. Linuron and metribuzin herbicides showed a high biodegradability during the composting process of green waste and sewage sludge decreasing their concentration up to 99% (Fountoulakis et al. 2010). The increasing use of household food waste (HFW) composting as potential waste management has also been a focus point for research since the lack of a proper composting technology may impact the final product quality involving pesticide presence. Růžičková et al. (2021) detected several pesticides such as methyl trithion, bifenox, dioxacarb, and desmetryn at concentrations between 0.3 and 16.3 µg kg^{-1} in compost from HFW. This indicates that the food leftovers contain pesticide residues which may end up in the final compost.

Pharmaceuticals and personal care products

Compounds of emerging concerns (CECs), such as pharmaceutical and personal care products (PPCPs), have ubiquitously been found in treated waste water and biosolids. Due to the extensive use of PPCPs, residues of these compounds may end up in the soil used for cultivating crops, and the derived food products. There are three major sources through which PPCPs end up in the environment: release during PPCP production, via wastewater containing pharmaceuticals excreted by humans, and via application of effluent or biosolids containing PPCPs (Gworek et al. 2021). The presence of personal care products in agricultural soils usually originates from irrigation with treated waste water. Pharmaceutical compounds, such as antibiotics (human and veterinary) and anti-inflammatory agents, can be present due to application of both treated waste water and animal or human waste (Gworek et al. 2021; Tasho and Cho 2016; Wu et al. 2015). Veterinary antibiotics are employed in livestock farming to control diseases and outside Europe they can also be used to promote animal growth. A large fraction of these compounds is not absorbed by animals after administration and is excreted unaltered via urine and feces. They are transferred to agriculture and horticulture through wastewater and the use of animal manure. Since animal manure is one of the most frequently used organic fertilizers in agriculture and horticulture, the monitoring, evaluation and control of veterinary antibiotics is of great importance to prevent food safety risks. Experimental studies have shown that the main antibiotics found in compost and animal manure are tetracyclines, sulfonamides, and fluoroquinolones (Berendsen et al. 2015; Zhang et al. 2015; Zhang et al. 2016). Among the organic wastes, animal manures present higher antibiotics concentrations than compost samples. Oxytetracycline followed by tetracycline were the most abundant compounds in manures, and fluoroquinolones was reported most in compost (Zhang et al. 2015). Regarding the evaluated different manures (pig, chicken, dugs, and cattle), authors showed that pig manures contained highest levels of antibiotics residues, probably due to the greater use of these compounds within these animals (Zhang et al. 2015).

Experiments for the determination of PPCPs in different long-term amended fields with several organic waste products showed that antibiotics (e.g. fluoroquinolones, carbamazepine) were mainly detected in sludge and manure, whereas anti-inflammatory compounds (e.g. ibuprofen) were primarily found in composted urban wastes (Bourdat-Deschamps et al. 2017). McClellan and Halden (2010) analyzed 110 biosolid samples and reported that 38 of the 72 analyzed PPCPs were present in concentrations ranging from 0.002 to 48 mg kg^{-1}dry weight, with triclocarban and triclosan as the most abundant ones.

When present in the soil, PPCPs can subsequently be transferred to crops, although experiments showed that translocation of these substances in plants is low. Compared to personal care products, antibiotics and other pharmaceuticals are more easily taken up by plants. Non-steroidal anti-inflammatory drugs (NSAIDs) are a group of pharmaceuticals that are less likely to occur in plants as these are primarily trapped in the soil matrix (Pullagurala et al. 2018). Uptake of residues from the soil by plants thus depends on the substance characteristics, such as persistence and mobility (Berendsen et al. 2018; Gworek et al. 2021). Berendsen et al. (2021) developed a strategy that determine a half-life value and mobility of antimicrobial active substances such as tetracyclines, quinolones, sulfonamides, macrolides and lincosamides. Authors remark that in these studies, not only the native substance should be targeted but also their degradation products since they might trigger similar effects as was demonstrated for tylosin. The plant species also plays a role, with leafy vegetables having the highest potential for CEC accumulation followed by root vegetables, cereals and fodder crops (such as maize and wheat), and fruit vegetables (Mansilla et al. 2021). Furthermore, soil characteristics impact plant uptake of contaminants (Pullagurala et al. 2018).

Prosser and Sibley (2015) assessed the human health risk related to the presence of PPCPs in plant tissue. They pointed out that, although PPCPs were detected in biosolid or manure amended soils, the toxically individual risks of PPCPs (based on the estimated hazard quotient) when applied to several crops were low presenting a minimum risk to human health. Nevertheless, the mixture of PPCPs could potentially present a food safety hazard and thus, there is a need to further investigate this field.

Hazards related to the use of edible resources in agriculture and horticulture

Crop residues, or plant by-products, are used to improve the soil quality by increasing the soil organic matter (SOM) as well as the macro and micro nutrients in the soil. The majority of the crop residues originate from cereals (Sarkar et al. 2020; Singh et al. 2015). To a lesser extent, residues are obtained from sugar crops, legumes, tubers, and oilseeds (Sarkar et al. 2020). These crop residues may be applied as fertilizer by directly adding them to the soil or indirectly by using them as raw material to produce biochar or compost. Crop residues may contain chemical food safety hazards as crops are known to take up substances during cultivation. These hazards may be present in the soil (such as heavy metals), the irrigation water (such as pharmaceuticals) or may originate from the air (e.g. polycyclic aromatic hydrocarbons (PAHs)). Furthermore, pesticides may be applied during crop cultivation of which residues may end up in or on the crop. Depending on the cultivar, chemical hazards might transfer and translocate throughout the plant. The following sections indicate the possible chemical food safety hazards related to the application of crop residues in agriculture and horticulture. Although pharmaceuticals were found to be taken up by plants (see section "Pharmaceuticals and personal care products"), no information was found on the possible accumulation of these substances when applied as crop residues in agriculture and horticulture.

Heavy metals and other elements

Various papers are available describing the application of biochar as a low-cost measure to remediate polluted soil as heavy metals will bind to the biochar (see e.g. reviews by (Chen et al. 2018; O'Connor et al. 2018; Singh et al. 2015; Wang et al. 2020). By binding of the heavy metals, biochar is capable of reducing the heavy metal content in the soil and its subsequent uptake in the plant. The effectiveness, however, depends on various factors, that is, the application time, site-specific factors, biochar characteristics, plant species and hazard characteristics (Chen et al. 2018; O'Connor et al. 2018). Apart from biochar, compost derived from crop residues has also been found to reduce heavy metal contents in crops grown on contaminated fields. Although compost in itself may contain heavy metals, levels in compost derived from green waste are low compared to sludge from wastewater treatment plants. During composting, the speciation of the metals changes leading to more fixed forms (Lopes et al. 2012; Thakali and MacRae 2021). Compost can then be useful for the remediation of contaminated soils (Lopes et al. 2012). Adejumo, Ogundiran, and Togun (2018), for example, showed that compost derived from sunflower and cassava reduced Pb, Cd and Cr levels in maize plants grown on lead-acid battery wastes polluted sites. Inonu et al. (2020) found that compost from palm empty fruit bunch (a by-product of palm oil production) reduced the metal (Aluminum (Al), Pb, and Cu) uptake in eggplant when cultivated in a post-tin mining area. The use of fresh solid olive husk (a by-product of the olive oil industry) showed to increase Manganese (Mn) bioavailability, while compost of the same by-product reduced the availability and plant uptake (De la Fuente et al. 2011). In contrast, Zhu, Zhong, and Wu (2016) showed an increase in methylmercury levels in rice grain when grown on paddy soils with (composted) rice straw.

The kind of metal, crop and soil characteristics influence the uptake of heavy metals and other elements by the crop (Peralta-Videa et al. 2009). The metal bioavailability to plants primarily depends on the soil pH with greater bioavailability at lower pH (Lopes et al. 2012). Some crops have shown to accumulate heavy metals in the edible parts of the plant. For example, Cd levels were found above the EU maximum limit (European Commission 2006a) for various leafy vegetables, fruiting vegetables tubers and root vegetables. Heavy metals, in general, tend to accumulate in the aboveground parts rather than in the roots of the crop (Hoffmans, Hoek-van den Hil, and van Asselt 2020; Nijkamp et al. 2017). When crop residues are used directly as soil fertilizers, this may lead to accumulation in crops subsequently cultivated on the amended soil.

Organic contaminants

During biochar production, the crop residues follow a heat treatment, pyrolysis, which may result in the formation of PAHs (De la Rosa et al. 2019; Downie et al. 2012; Ruzickova et al. 2021; Singh et al. 2015), PCDD/Fs, heavy metals, PCBs, chlorophenols (CPs), and chlorobenzenes (CBzs) (Downie et al. 2012). PAHs are frequently mentioned as a potential problem arising during biochar production. Levels of PAHs found in biochar depend on the crop residue used, which may be quite diverse, for example, wheat (Bian et al. 2018), rice (Liu et al. 2021), or sugar cane (Puga et al. 2015). However, the biochar production process, i.e. the applied temperature and reactor used, has a larger impact on the PAH formation than the crop residue used (De la Rosa et al. 2019). Apart from formation of chemical hazards during biochar production, various organic contaminants, such as PAHs, PCBs, dioxins and PFAS, may also be present in the soil due to industrial activities. The application of compost or biochar as soil fertilizer may further introduce chemical hazards in the soil. The characteristics of the compounds that are present influence the uptake and translocation in the plant (Ruzickova et al. 2021; Singh et al. 2015). For example, the hydrophobicity of PFAS plays a significant role in plant uptake (Pullagurala et al. 2018). Besides compound characteristics, soil characteristics, crop characteristics (such as lipid content) and the growth cycle of the crop can also influence plant uptake. For some hazards, like PCBs and PAHs, the levels found in crops are low or negligible (Lopes et al. 2012; Paris et al. 2018). In general, compost prepared from green waste shows low levels of organic contaminants (Lopes et al. 2012).

Plastics and nanomaterials

The environmental contamination with different sized plastics has increased due to the widespread plastic production and use (Li et al. 2020). Engineered nanomaterials (ENN) may also end up in the soil when they are disposed in for example landfills. Experimental studies have shown that agricultural crops are capable of taking up microplastics (Li et al. 2020) and nanomaterials. The concentration in the soil and the crop type influence the levels found in the crops (Pullagurala et al. 2018). Plastics tend to accumulate in the roots of the plants. Root vegetables such as carrots and radish may thus be at increased risk. Higher temperatures and reduced humidity have been shown to increase accumulation in the crop (Li et al. 2020). Although plastics initially accumulate in the roots of the plant, they may translocate to other parts of the plants (Li et al. 2021). More research is needed on the accumulation and translocation of micro and nano plastics into the crops, especially when reused in agriculture and horticulture (Pullagurala et al. 2018).

Pesticides

Organochlorine pesticides (OCPs), such as chlordane and DDT, are persistent in the environment due to their frequent use as plant protection products in the past and can be taken up by agricultural crops. However, pesticide residues found in crops are primarily due to the direct application on the plant rather than via soil uptake (Pullagurala et al. 2018). Indeed, pesticide residues are frequently found in crops and sometimes above the EU maximum residue limits (MRLs) (Banach et al. 2019; Hobe et al. 2020; Hoffmans, Hoek-van den Hil, and van Asselt 2020; Klüche, Hoek-van den Hil, and van Asselt 2020; Nijkamp et al. 2017). No information was found on the effect of reusing crop residues as fertilizers in soil on the pesticide levels.

Natural toxins

Some crops contain plant toxins or mycotoxins produced by fungi. Legumes, such as lupine, contain quinolizidine alkaloids. When present in the soil, these compounds proved to be rather stable (Hama and Strobel 2020). Potatoes may produce glycoalkaloids and tropane alkaloids, the levels of which depend on the crop species and climatic conditions (Nijkamp et al. 2017). Crops may also produce (poly)phenols, which may have biological effects. However, since these compounds are not very persistent in the soil, they are unlikely to enter the food chain and impact human health when crop residues are applied as fertilizer (Goss, Tubeileh, and Goorahoo 2013). Fungal contamination of cereals has shown to result in the presence of mycotoxins such as deoxynivalenol and aflatoxins depending on climatic conditions (Klüche, Hoek-van den Hil, and van Asselt 2020). Cereal by-products, such as husks and fibers usually contain higher levels of mycotoxins than the grains. These by-products are currently primarily used in animal feed (see section "Natural toxins") and no information was found on the reuse in agriculture and horticulture.

Hazards related to the use of edible resources in feed

Both plant and animal by-products may be reused in animal feed production in order to avoid wasting resources. As compared to the main products intended for human consumption, higher concentrations of chemical hazards, or even other chemical hazards, may be seen in the by-products. An overview of known chemical hazards related to these resources is given below.

Heavy metals and other elements

Heavy metals may accumulate in sugar beet. Sugar beet pulp, a by-product from the sugar industry, which is frequently used as feed ingredient, can, therefore, contain heavy metals. As indicated previously, heavy metals are primarily found in leafy vegetables, tubers and root vegetables. Potatoes may thus also contain heavy metals with higher concentrations in the peel than in the potato itself. Potato peels may be used as animal feed. Cereals and seeds usually contain low levels of heavy metals. Residual products from bakeries that can be used in animal feed are thus not expected to contain high concentrations of heavy metals (van der Fels-Klerx et al. 2019).

Fish meal, mostly derived from fish, or fish parts with a low economic value are frequently contaminated with heavy metals, such as Cd, or mercury (Hg) (Adamse, Van der Fels-Klerx, and de Jong 2017). As a result, fish meal or other fish by-products are a major source of heavy metals in the feed supply chain (van der Fels-Klerx et al. 2019).

In aquaculture, by-products from the food industry, processed into pellets are investigated as fish feed. The heavy metal content of food waste-based feed pellets with cereals (53% cereals, 10% fruit and vegetables, 8% bone meal, 29% non-food waste products: corn starch, fish meal, other) and food waste-based with cereals and meat products (28% cereals, 25% meat products, 10% fruit and vegetables, 8% bone meal and 29% non-food waste products) were compared to traditional fish meal. Food waste-based feed pellet, with or without meat, showed much higher concentrations (more than three times higher) of heavy metals, especially Arsenic (As), Chromium (Cr), Cu, Ni and Zn, than traditional fish meal. The highest concentrations of Cu and Zn were found in the pellets containing meat products. As a result, the farmed fish itself also contained higher heavy metal concentrations when fed with food-waste based pellets than when fed with fish meal (Cheng et al. 2016; Cheng et al. 2014).

Organic contaminants

Fish oil or fish meal, used as ingredients in animal feed, are two products frequently contaminated with dioxins and PCBs due to contamination via the environment (Ortiz et al. 2011; Suominen et al. 2011; van der Fels-Klerx et al. 2019). Other ingredients, which are subject to a drying process may also contain dioxins and PCBs. When contaminated fuels such as non-feed grade oil or waste wood, or wrong techniques are used during the process, dioxins and PCBs can be introduced into the product. This may be the case for old bread, which after drying and processing can be reused in animal feed. Citrus pulp, before being used in animal feed, is frequently mixed with clay, possibly leading to dioxin or PCB contaminated feed (Hoogenboom et al. 2004; van der Fels-Klerx et al. 2019).

Brominated flame retardants (BFRs), mainly PBDEs and HBCDs, are fat soluble and can be found in fish oil used in animal feed (Ortiz et al. 2011). Of all food and feed products analyzed in the UK in 2013, fish feed, consisting of mainly fish meal and fish oil, contained the highest concentrations PBDE and HBCD (Fernandes et al. 2016).

PFAS, including PFOA and perfluorooctane sulfonic acid (PFOS), can be found in edible resources in feed. PFOA has been found in the environment due to contaminated biosolids or manure applied as fertilizer, (Kowalczyk et al. 2013; Lupton et al. 2012), and illegal waste disposals or contaminated river water (Kowalczyk et al. 2013; Zafeiraki, Vassiliadou, et al. 2016). It has been shown that PFAS can be taken up from the soil by maize (Krippner et al. 2015) and other feed crops like grass. PFOA, when present in animal feed, seems to be largely absorbed and excreted in the animals' urine, as shown in experiments performed on Angus cattle and sheep (Kowalczyk et al. 2013; Lupton et al. 2012). PFOS on the other hand, accumulates in organs like liver and kidney. PFOS binds to protein albumin, mainly present in the blood, liver and eggs (De Vos et al. 2008). An experiment performed on dairy sheep showed that plasma levels of PFOS increased continuously, and a limited amount was excreted via feces (4–5%) and via milk (2%) (Kowalczyk et al. 2013). PFOS has been detected in eggs in China, the Netherlands and in Greece (Wang et al. 2008; Zafeiraki, Costopoulou, et al. 2016). PFAS are also detected in variable concentrations in fishmeal (Suominen et al. 2011).

Finally, PAH's can end up in animal feed via by-products containing fats and fatty acids, such as vegetable or fish oils, but can also arise during the processing of by-products (e.g. drying) (van der Fels-Klerx et al. 2019). However, most agricultural animals have the ability to metabolize and excrete PAHs, and for that reason they hardly accumulate in animal products through contamination of the feed.

Pesticides and pharmaceuticals

Pesticide residues are frequently found in crops and, in this way, can be present in plant-based by-products. One example is the pesticide paraquat used in the production of soybean outside the European Union. Paraquat does not dissolve in the soybean oil, the main product, but remains in the soy meal, the by-product used in animal feed (van der Fels-Klerx et al. 2019).

Pharmaceutical residues might be present in feathers, hairs, bone meal, egg meal, eggshells, and fish meal. These by-products could potentially be a source of re-introduction of pharmaceutical residues in the supply chain when incorporated in animal feed. Furthermore, during the production of bioethanol, it is not uncommon to add bacterial growth inhibitors such as antibiotics to optimize the fermentation process. Residues of these antibiotics might be present in the by-product distiller's dried grains with soluble (DDGS), used in animal feed (van der Fels-Klerx et al. 2019).

Natural toxins

Ricinus communis, commonly called the castor bean or castor oil plant, is one of the most important oilseed crops in arid and semi-arid regions. Castor bean meal is the left-over product of the biofuel industry. The plant is, however, not (directly) edible and is even listed as an undesirable ingredient in animal feed in the Annex of Directive 2002/32/EC due to inherent toxins, including ricin (European Commission 2002). Ricin is one of the most poisonous naturally-occurring substances known. Multiple detoxification strategies have been studied to produce detoxified castor meal. Even though ricin levels were significantly reduced in the detoxified castor meal, this by-product still induced negative effects such as a reduced growth in pigs, poultry and cattle (Akande, Odunsi, and Akinfala 2016).

Another example of a widely grown crop is cotton. The by-product, cotton seed meal, can be used as animal feed. However, its use in animal feed is limited due to, amongst others, the presence of gossypol. Cottonseed meal can undergo fermentation to lower the concentration of gossypol. However, the feeding of high quantities of fermented cottonseed meal still led to immunotoxin and hepatotoxic effects in broilers (Xiong et al. 2016).

Mycotoxins are also a potential hazard in by-products. Two by-products at risk of mycotoxin contamination and used as animal feed ingredient are DDGS, a by-product from the bio-ethanol industry, and germ, rootlets or brewer's spent grains, which are by-products from the beer industry. Mycotoxins mainly accumulate in the outer fractions of the grain, such as the fibers and husks. Germ and rootlets tend to contain the highest levels of deoxynivalenol. Deoxynivalenol concentration in spent grains seem to be highly variable (Mastanjević et al. 2019). Regarding DDGS, often three times the concentration of mycotoxins is found in DDGS compared to the raw material (van der Fels-Klerx et al. 2019).

Ryegrass intended for turf (tall fescue, perennial ryegrass) is frequently infected with an endophyte fungus (*Neotyphodium spp.*) that produces mycotoxins. The relationship between the fungus and the grass is symbiotic: the grass provides nutrients to the fungus and the fungus produces toxins that helps the grass to defend itself against insects, diseases and grazing animals. Traditionally in the US, the seed is harvested in July or August and the straw residue is burnt in August. Alternative uses of the straw are being investigated. The straw is, for example, currently used as animal feed, especially in Asia. However, the endophyte fungus which can be present in straw produces toxic alkaloids, including several ergot alkaloids (e.g. ergovaline) and lotitrem alkaloids (e.g. lotitrem B) (Craig 2015).

Conclusion

This literature review demonstrated the potential presence of chemical food safety hazards when using non-edible resources in agriculture and horticulture as soil amendments since both urban and animal wastes showed the presence of many pollutants. It can be said that the predominant hazards in these organic resources are heavy metals, human and veterinary pharmaceutical and personal care products. Although poultry manure increases soil organic matter, it is a potential source of veterinary medicine residues in soil due to the extended use of these chemicals in poultry production. Biosolids can cause the presence of pharmaceuticals and personal care products in agricultural soil and due to frequent application may result in a worrying accumulation affecting plant uptake. Most information regarding the hazards present in sewage sludge was related to heavy metals and organic contaminants originating from industrial activities that require higher control to avoid soil contamination. Therefore, there is a need to develop a low-cost, environmentally friendly recycling technology to remove metals from industrial sludge and thus be able to use the sludge as fertilizer without posing food safety problems. It should be mentioned that some of the metals found, such as Zn, Ni and Cu, are also essential elements that are – to certain concentrations – beneficial for plant and human health. However, elevated levels of these elements may lead to adverse health effects. Further research is needed to establish the relationship between levels in the soil and levels in the crops in order to determine safe levels for application of sludge and manure in agriculture and horticulture. Recent developments in the process of waste composting indicate that it seems a way forward in reusing organic wastes such that it reduces the presence of some chemical hazards. Further research is needed in order to achieve a high-quality compost that minimizes the presence of food safety hazards.

When considering the use of crop residues in agriculture and horticulture, the literature review revealed that many chemical hazards may be taken up by plants and can be translocated to the edible parts of the plant. Most information was available on the application of biochar as remediation of contaminated soil. However, limited information was available on chemical hazards related to the reuse of crop residues in agriculture and horticulture either through direct application in the soil or indirectly through the use of compost or biochar derived from crop residues. Although the application of compost and biochar tends to decrease hazard levels in the soil and subsequent uptake by crops, the biochar production process is susceptible for the production of PAHs. The production process needs to be steered such that PAH production is limited. When crop residues are directly applied as soil fertilizer, chemical hazards present in the crops may accumulate in the soil and crops grown on this soil. Main hazards related to crop residues are heavy metals and pesticides. For cereals, additionally, mycotoxins are a relevant group of chemical hazards. Plant toxins are relevant for legumes and potatoes. The extent to which these hazards accumulate when crop residues are reused in agriculture and horticulture is largely unknown. Therefore, further research is needed to establish levels of food safety hazards in crop residues, their persistence and accumulation in the soil and subsequent transfer in the crop. Based on such research, counter measures can be taken allowing the safe application of crop residues in agriculture and horticulture.

Finally, very few recent references dealt with chemical hazards in by-products used as animal feed. The literature showed that a variety of chemical hazards can be present in both plant- and animal-based by-products. Most information was available for fish meal and oil, potentially containing high levels of dioxins, PCB's, BFR's, PFAS, and heavy metals. In general, chemical hazards, such as natural toxins, might be present at higher levels in the by-products than in the raw product, as has been demonstrated for DDGS or brewer's spent grains. Other examples of by-products potentially containing high levels of chemical hazards are feathers, or eggs from laying hens that are treated with antibiotics, which are not used for human consumption but may be used for animal feed.

Overall, chemical hazards may be present in the resources used to close the loop in agriculture. Depending on the origin of the resources, different compounds may be found. Further research is needed to ensure the safe reuse of resources. It is recommended to identify potential hazards at an early stage and take precautionary measures to prevent accumulation in the food supply chain when moving toward a more circular agriculture. This can be achieved by performing more analyses on resources used in agriculture and horticulture and feed and/or using a modeling approach to predict the fate of chemical hazards in the food supply chain when closing the loop.

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Annex.Search terms used for the literature review

1. *Chemical hazards related to non-edible sources in agriculture and horticulture*

1 Commodity (in TITLE):

"Sewage sludge" OR sludge* OR sewage* OR sediment* OR slime* OR grease* OR "sewer waste" OR struvite* OR kaolite* OR sepiolite* OR bentonite* OR mineral* OR slurry* OR "Organic waste" OR "organic household waste*" OR "biological waste*" OR "biological trash*" OR "organic debris*" OR "organic remain*" OR "organic trash*" OR "biological remain*" OR swill* OR "garden waste*" OR "roadside waste*" OR "roadside clippings" OR grass* OR "hedge clippings*" OR "organic material" OR "green waste" OR weed* OR hedge* OR hay* OR roughage* OR silage* OR straw* OR molasse* OR pulp* OR manure* OR faeces* OR dung* OR compost* OR droppings* OR excreta* OR excrement* OR "blood meal*" OR "hemoglobin powder*" OR "haemoglobin powder*" OR "Feed cake*"

AND #2 Side streams (in TITLE):by-product* OR byproduct* OR co-product* OR coproduct* OR waste* OR "side stream*" OR "rest stream*" OR residue*

AND #3 Application (in TITLE-ABS-KEY):circular* OR feed* OR forage* OR roughage* OR fertiliser* OR fertilizer* OR compost* OR champost* OR biochar* OR biosolid* OR "green waste*" OR "soil application" OR "sustainable agriculture"

AND #4 Chemical hazards (in TITLE-ABS-KEY):pollutant* OR "chemical hazard*" OR hazard* OR toxin* OR "agricultural chemical*" OR agrochemical* OR "chemical compound*" OR "chemical substance*" OR residu* or pesticide* or "plant protection product*" or organophosphate* or organochlorine* or carbamate* or pyrethroid* or "heavy metal*" or element* or "mineral oil*" or mycotoxin* or pharmaceutical* or "vet* drug*" or "vet* medic*" or antibiotic* or antimicrobial* or antiparasitic* or anti-parasitic* or NSAID* or sedative* or hormone* or steroid* or "beta*agonist*" or "plant toxin*" or "Pyrrolizidine alkaloid*" or glycoalkaloid* or radionuclide* or dioxin* or PCB* or perfluor* or PFAS or "flame retardant*" or PAH* or "polycyclic aromatic hydrocarbon*"

AND #5 Human health (in TITLE):

"public health" OR HACCP OR "consumer protection" OR "food safety" OR "risk assessment*" OR "risk analys*" OR hazard* OR "human health*" OR "health impact" OR "health risk*" or uptake* or *transfer* or accumulation* or concern or Toxic*)

AND PUBYEAR AFT 2010

AND NOT #6: Exclusion terms (in Title):nutrient* OR nitrog* OR phosphor* OR lithi* OR *nitrat* OR "zinc" OR "element*" OR gene* OR bacteri*

2. *Chemical hazards related to crop residues in agriculture, horticulture and feed*

#1 Commodity (in TITLE):crop* or plant* or Cereal* or grain* or barley* or maize* or corn* or millet* or oat* or rice* or rye* or wheat* or buckwheat* or sorghum* or *seed* or cocoa* or cotton* or groundnut* or olive* or palm* or soy or lucerne* or flax or legume* OR pulse OR pulses OR bean* OR pea OR peas OR cowpea* OR chickpea* OR lentil* OR bambara* OR lupin* OR soy* or asparagus OR cardoon* OR celeriac* OR garlic* OR fennel* OR kohlrabi* OR kurrat* OR leek* OR lotus* OR nopal* OR onion* OR shallot* OR carrot* OR ahipa* OR arracacha* OR "bamboo shoot*" OR beetroot* OR "beet root*" OR gobo OR burdock* OR cassava* OR manioc* OR chufa* OR "tigernut tuber*" OR daikon* OR mooli* OR ginger* OR "root* parsley*" OR "parsley root*" OR horseradish* OR jicama* OR parsnip* OR radish* OR rutabaga* OR swede* OR salsif* OR scorzonera OR skirret* OR potato* OR taro* OR turnip* OR ulluc* OR "water chestnut*" OR wasabi* OR yacón OR yacon OR yam* OR rhubarb* OR "pie plant" OR samphire* OR nuts or almond* or chestnut* or hazelnut* or groundnut* or "pine nut*" or pistachio* or walnut* or cashew* or coconut* or "macadamia nut*" or peanut* or pecan* or tomato* OR aubergine* OR pepper* OR courgette* OR zucchini* OR cucumber* OR cucurbit* OR gourd* OR pumpkin* OR squash* OR kabocha OR hokkaido OR tinda OR eggplant* OR egg*plant OR chilli* OR chili* OR oliv* OR okra or fruit* or apple* or citrus* or fig* or grape* or orange* or apricot* or strawber* or banana* or kiwi* or mango* or pear* or brocco* OR cauliflower* OR sprout* OR cabbage* OR chicory OR spinach* OR "turnip top*" OR "turnip green*" OR kale OR chard OR lettuce* OR endive OR escarole* OR salad OR choi OR choy OR artichoke OR arugula OR "beet green" OR bitterleaf OR celery OR celtuce OR "collard green*" OR *cress* OR epazote OR "garden rocket" OR komatsuna OR "mizuna greens" OR "mustard green*" OR "leaf mustard*" OR radicchio OR rapini OR tatsoi OR chaya OR chickweed OR "Chinese mallow" OR Chrysanthemum OR "fat hen" OR "fluted pumpkin" OR samphire OR "Greater plantain" OR "jute plant" OR Karkalla OR "Lagos bologi" or orache OR purslane OR rucola OR sculpit OR stridolo OR soko OR "spleen amaranth" or "leafy green*"

AND #2 Side streams (in TITLE):by-product* or byproduct* or co-product* or coproduct* or hull* or fibre* or leaves or leaf* or straw* or stalk* or bran* or middling* or shell* or flake* or husk* or pulp* or peel* or waste* or "side stream*" or "rest stream*" or residue* or foliage* or mash* or meal* or oil*

AND #3 Application (in TITLE-abs-key):

Circular* or Feed* or forage* or roughage* or "soil application*" or "sustainable agriculture" or fertiliser* or fertilizer* or compost* or champost* or biochar* or biosolid* or "green manure*" or "green waste*" or "sustainable agriculture"

AND #4 Chemical hazards (in TITLE-ABS-KEY):pollutant* OR "chemical hazard*" OR hazard* OR toxin* OR "agricultural chemical*" OR agrochemical* OR "chemical compound*" OR "chemical substance*" OR residu* or pesticide* or "plant protection product*" or organophosphate* or organochlorine* or carbamate* or pyrethroid* or "heavy metal*" or element* or "mineral oil*" or mycotoxin* or pharmaceutical* or "vet* drug*" or "vet* medic*" or antibiotic* or antimicrobial* or antiparasitic* or anti-parasitic* or NSAID* or sedative* or hormone* or steroid* or "beta*agonist*" or "plant toxin*" or "Pyrrolizidine alkaloid*" or glycoalkaloid* or radionuclide* or dioxin* or PCB* or perfluor* or PFAS or "flame retardant*" or PAH* or "polycyclic aromatic hydrocarbon*"

AND #5 Human health (in TITLE):

"public health" OR HACCP OR "consumer protection" OR "food safety" OR "risk assessment*" OR "risk analys*" OR hazard* OR "human health*" OR "health impact" OR "health risk*" or uptake* or *transfer* or accumulation* or concern or Toxic*)

AND PUBYEAR > 2010

AND NOT #6 Exclusion terms (in Title):

wastewater* or irrigation* or sludge*

3. *Chemical hazards related to animal by-products in feed*

#1 Commodity (in TITLE-ABS-KEY):

Dairy* or milk* or butter* or cheese* or caseinate* or lactose* or whey or "Animal fat*" or "animal protein*" or gelatine* or blood* or "catering reflux*" or feather* or "slaughter*" or egg* or "fishery product*" OR fish* OR "common sole" or pangasius OR tilapia OR perch OR eel* or herring* or "sea bass*" or seabass* or anchovy* or shellfish OR crustacean* OR mollus* or shrimp* or prawn* or crab* or lobster* or squid* or octopus or haddock* or monkfish* or anglerfish* or mussel* or clam* or salmon* or oyster* or plaice* or ray* or turbot*or tuna* or mackerel* or trout*

AND #2 Side streams (in TITLE-ABS-KEY):by-product* or byproduct* or co-product* or coproduct* or hull* or fibre* or shell* or flake* or husk* or pulp* or peel* or waste* or "side stream*" or "rest stream*" or residue* or mash* or meal* or oil*)

AND application (in TITLE-ABS-KEY):circular* OR feed* OR forage* OR roughage* OR fertiliser* OR fertilizer* OR compost* OR champost* OR biochar* OR biosolid* OR "green waste*" OR "soil application" OR "sustainable agriculture")

AND #3 Chemical hazards (in TITLE-ABS-KEY):pollutant* OR "chemical hazard*" OR hazard* OR toxin* OR "agricultural chemical*" OR agrochemical* OR "chemical compound*" OR "chemical substance*" OR residu* or pesticide* or "plant protection product*" or organophosphate* or organochlorine* or carbamate* or pyrethroid* or "heavy metal*" or element* or "mineral oil*" or mycotoxin* or pharmaceutical* or "vet* drug*" or "vet* medic*" or antibiotic* or antimicrobial* or antiparasitic* or anti-parasitic* or NSAID* or sedative* or hormone* or steroid* or "beta*agonist*" or "plant toxin*" or "Pyrrolizidine alkaloid*" or glycoalkaloid* or radionuclide* or dioxin* or PCB* or perfluor* or PFAS or "flame retardant*" or PAH* or "polycyclic aromatic hydrocarbon*"

AND #4 Human health (in TITLE):

"public health" OR HACCP OR "consumer protection" OR "food safety" OR "risk assessment*" OR "risk analys*" OR hazard* OR "human health*" OR "health impact" OR "health risk*" or uptake* or *transfer* or accumulation* or concern or Toxic*

AND PUBYEAR aft 2010